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TALL OIL

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SUMMARY

Description

Tall oil is a pale amber to dark brown oily liquid derived from wood; it has a slightly resinous, slightly fishy odor. Physical properties and chemical composition vary considerably according to the source of the wood and certain processing methods. The main constitutents are fatty acids (35-60%), resin acids (35-55%), and unsaponifiable matter (5-10%). Tall oil is combustible. The flash point is 360°C.

Occurrence

Tall oil is a by-product of the kraft or sulfate wood pulp industry. The highly alkaline pulping chemicals convert the wood fats and resins to soaps. Tall oil is obtained when the soaps (skimmings) are acidified. Various species of pine trees are the richest source of tall oil. The yield varies from 60 to 110 pounds per ton of pulp (161). Tall oil and its products have many commercial applications including: chewing gum bases, paper and paperboard products, emulsifiers, anti-foam agents, animal feeds, paper sizes, water-proofing agents, cellulose, oil cloth and linoleum, synthetic rubber, wetting agents, adhesives, binders, soaps, disinfectants, fungicides, insecticides, flotation agents, and coatings (161).

Acute Toxicity

No reports on the toxicity of whole tall oil have been found. The oral LD_{50} for refined tall oil resin is 7600 mg/kg for rats and 4600 mg/kg for mice and guinea pigs (177). A group of commercial epoxidized tall oil products used in coatings have oral LD_{50s} ranging from 10,000 to 54,000 mg/kg in rats and 16,000 to > 20,000 mg/kg in rabbits (158). The minimum lethal concentration of crude tall oil soap and the sodium salts of its fatty acid fraction for fish is 5 ppm. The MLC of the resin acids fraction sodium salts is 1 ppm (152).

Short-term Studies Tall oil fatty acid distillate, fed at a level of 35% in the diet of rats for four weeks, resulted in 100% fatalities. The mortality rate of the ethyl esters was approximately 70% and that of the glycerides 50%. At the 13% dietary level, the mortality rate was only 0-5%, but growth was retarded. Hydrogenated tall oil fatty acid glyceride margarine, on the other hand, caused total fatalities of less than 5% and growth rates approximated those of the controls (132).

Refined tall oil rosin, at a level of 5% in the diet of rats, killed all of the animals within two weeks. At a concentration of 1%, no fatalities resulted, but initial growth rates were depressed and liver enlargement resulted after 90 days. A dietary resin content of 0.2% was not deleterious (177). In young beagle dogs, liver enlargement resulted also at the 1% dietary level, but not at a concentration of 0.05%. No histologic abnormalities were detected in any of the organs, including the liver, in either study (177).

Crude tall oil, refined tall oil, and tall oil resins fed to chicks at a level of 5% in the diet retarded growth. The growth retarding factor seemed to reside in the resin acids fraction (142).

Tall oil fatty acid esters in the diet of cattle caused a slight decrease in milk fat production at 4% but not at 3%. There were no other toxic effects at either level (015).

Human toxicity data are meager. Tall oil and the resins acids are slightly irritating to the skin and mucous membranes (126, 139). Abietic acid is also moderately toxic by ingestion. β-sitosterol causes anorexia, gastrointestinal cramps, and diarrhea in some patients on very large doses (112, 125).

Long-term Studies

Refined tall oil rosin, at a level of 1% in the diet of rats for a period of two years, depressed the growth rate slightly and caused liver enlargement unaccompanied by any histologic abnormalities. Other organs were not affected. Hematology, urinalysis, and liver and kidney function test values were within normal limits throughout. At dietary levels of 0.2% and 0.05% no adverse effects were noted. (177)

Rats fed tall oil glyceride margarine at a level of 13% in the diet had a much longer life span than control animals fed butter (132).

Special Studies

Oxidized oleic acid administered to albino mice parenterally caused chromosome abnormalities in bone marrow cells (111). Life-time carcinogenicity studies in mice with two epoxidized tall oil products were negative for tumorigenic potential (158). Refined tall oil rosin at 1% in the diet did not increase tumor incidence in dogs or rats over a period of two years (177). Tall oil margarine in the diet at levels of 13 and 35% had no detrimental effect on the reproductive capacity of rats (132).

Biochemistry

Tall oil undergoes oxidative changes on standing. Disproportionation, or reducing the number of double bonds in certain constituents, improves stability (014). Tall oil esters are absorbed well (93-96%) from the intestinal tract of rats (132). Feeding tall oil preparations as diet supplements to chicks, rats, and cattle resulted in changes in milk fat, egg yolk, and tissue lipids. Food consumption and growth were depressed when chicks were fed tall oil at a level of 5% in the diet (142). Yolk lipid changes involved marked increase in oleic acid and corresponding decreases in palmitic, stearic, and linoleic acids. The tall oil esters also decreased egg weight and impaired certain baking characteristics of the eggs (014).

The yield of milk fat was depressed by tall oil fatty ethyl esters at a level of 4% but not at 3% in the diet of cattle. The iodine number increased significantly at both levels (015).

Feeding tall oil fatty acids at levels of 13% in the diet of rats caused an increase of linoleic acid and a decrease of palmitic in adipose tissue. Plasma lipid changes involved an increase in oleic, and a decrease in linoleic and palmitic acids in the cholesterol ester fraction. The stearic acid content of liver triglycerides was higher in the tall oil group than in the soybean oil controls. More than 16% cis-5,9,12-octadecatrienoic acid was found in the fecal lipids (132).

Consumer Exposure

Consumer exposure to whole tall oil is limited mainly to certain food packaging and processing articles where it is used as a manufacturing additive. Consumer exposure to tall oil constituents and compounds of tall oil, on the other hand, is quite broad as a result of their widespread commercial applications. Glycerides prepared from refined tall oil fatty acids mixed with other oils has been sold as Grade B olive oil (113). Patents have been granted for the use of tall oil fatty acids in salad oils, shortenings, and margarine (132). Tall oil rosin preparations which meet certain specifications are used in synthetic chewing gum bases (009). A number of other tall oil compounds are used as emulsifiers, de-foaming agents, anti-caking preparations, surface coatings, etc. in various areas of the food industry (104). β-sitosterol from tall oil is used as an anti-cholesterolemic agent (112, 125,140). Sex hormones and other steroid medicinal products also are being prepared from tall oil sterols (006). It is predicted that the use of tall oil products will increase sharply in view of the continued growth of the paper industry, and the fact that tall oil is the cheapest source of fatty acids in the world (007,172).

Safe Limits

Abietic acid may be used in concentrations up to 0.0026% (26 ppm) as a carrier in enriched rice. Glyceryl abietate is used as an emulsifier in alcoholic beverages and fruit drinks in amounts up to 0.006%. Abietic and pimaric copolymers are permitted as surface coatings for fresh fruits up to a concentration of 0.02% (049). Potassium abietate is allowed in concentrations up to 1 ppm with fresh fruits and vegetables (011). Oleic acid is used in amounts ranging from 0.02 - 25 ppm in candy, baked goods, ice cream, ices, beverages. Mono- and diglycerides of fatty acids may be used in emulsifiers in concentrations up to 20 g/kg of emulsifier (031). Polyglycerol esters of fatty acids have acceptable daily intake values for man of 0 - 12.5 mg/kg (unconditional) and 12.5 - 25.0 mg/kg (conditional); the ADI of acetic and fatty acid esters of glycerol is 0 - 100 mg/kg (unconditional) (171).

CHEMICAL INFORMATION

I. Nomenclature

A. Common names: Tall oil[a] (Tallol; Liquid rosin)

B. Chemical name: Tall oil

C. Trade names[b]: Crude tall oil: Acintol C; Advanol; Campol (8485); Covoil; Opail; Plymouth; Superior; Trostol; Ligro

Distilled and refined tall oils: Acintol (D, etc.); Acolin; Aconon; Acosix; Campol (36-5, etc.); Facoils; Indusoil; Pamaks; Bogol; Rosoil; Unitol.

D. Chemical Abstracts Services Unique Registry Number: MX8002264

II. Empirical Formula

A. Tall oil. Tall oil is a mixture of fatty acids, resin acids, and unsaponifiable matter. It is obtained from pine wood as a by-product of the kraft or sulfate process in the manufacture of paper. The strong alkaline chemicals used for pulping (digesting) the wood convert the fats and resins present into soaps (sulfate soap) which is skimmed off the surface of the spent liquor. When the soaps or skimmings are acidified, tall oil results. The yield and composition vary with the species of pine, its geographic source, and the method of handling after the wood is cut. Normally, from 60 to 110 pounds of crude tall oil are obtained per ton (2000 lbs) of pine wood pulp (90% dry). In general, fatty acids range from 35-60%, resin[c] acids from 35-55%, and unsaponifiable matter from 5-10%.

B. Constituents

Fatty acids - Oleic acid or cis-9-octadecanoic acid ($C_{18}H_{34}O_2$) and linoleic acid or cis-9,12-octadecadienoic acid ($C_{19}H_{32}O_2$) are the main components. Seppanen (132) points out that the presence of cis-5,9,12-octadecatrienoic acid and cis-5,11,14-eicosatrienoic acid in tall oil makes it unique because the location of the double bonds in these acids differ from those in the octadecatrienoic and eicosatrienoic acids of other natural fats. Other fatty acids that have been found in tall oil are shown in Table 1.

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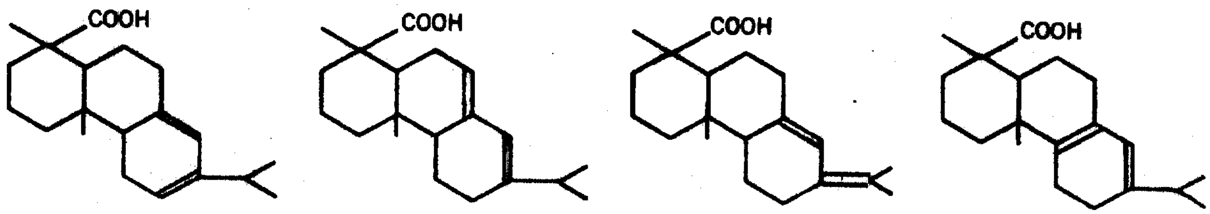
a "Tall" is the Swedish word for "pine". See Appendix for large list of synonyms.

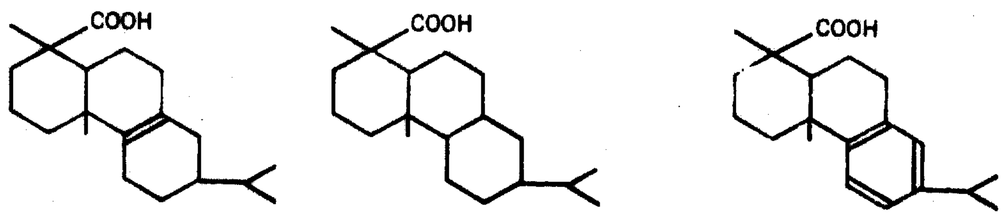
b Manufacturers are listed in Appendix.

c "Resin" and "rosin" are used synonymously in the tall oil literature and in this monograph.

ROSIN ACIDS · ABIETIC TYPES

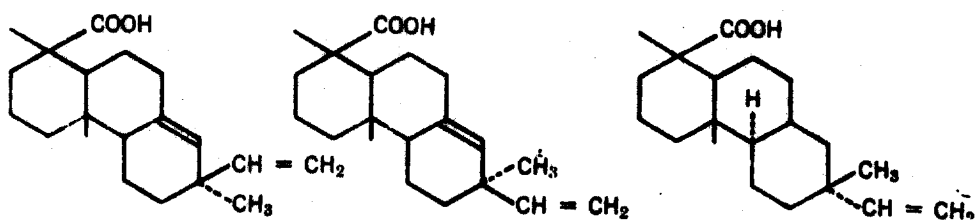


Levopimaric Acid | Abietic Acid | Neoabietic Acid | Palustric Acid

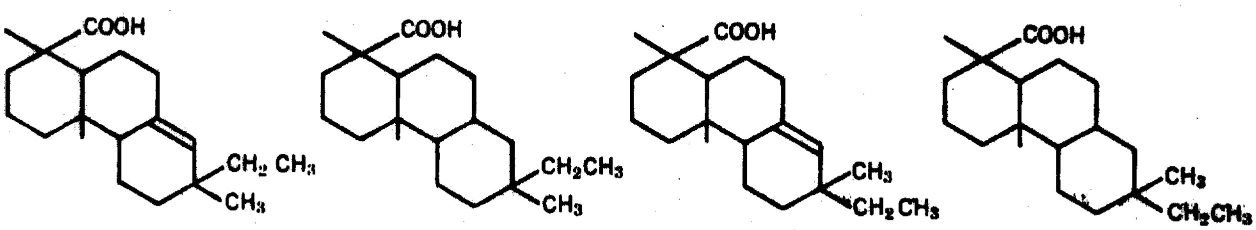


Dihydroabietic Acid | Tetrahydroabietic Acid | Dehydroabietic Acid

ROSIN ACIDS · PIMARIC TYPES



Pimaric Acid | Isopimaric Acid | Sandaracopimaric Acid



Dihydropimaric Acid | Tetrahydropimaric Acid | Dihydroisopimaric Acid | Tetrahydroisopimaric Acid

Fig. 1 Rosin Acids[a]

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[a] Courtesy of Arizona Chemical Company, Wayne, New Jersey

Unsaponifiable fraction - This fraction consists mainly of hydrocarbons and sterols, but its composition is exceedingly complex. β-sitosterol ($C_{29}H_{50}O$) is the major sterol; α-sitosterol ($C_{30}H_{50}O$) and stigmasterol ($C_{29}H_{48}O$) are also found. The hydrocarbon fraction contains sesqui- and diterpene hydrocarbons as well as aliphatic and other types. About one-third of the unsaponifiable fraction consists of diterpene alcohols and aldehydes of pimaric, isopimaric, abietic, and dehydroabietic types. Lignoceryl, aracinol, and behenic alcohols are present. In addition, trans-3,5-dimethylstilbene, "tall oil ketone", campesterol, β-sitostanol, squalene, cycloartenol, and pinosylvin dimethyl ether have been isolated as well as a number of minor components. Small amounts of highly-colored and highly-odorous materials are also present in tall oil. Current refining methods do not always eliminate all of these, such as certain sulfur compounds; this fact has barred the use of some tall oils in certain applications (172).

As indicated earlier, tall oils vary considerably in composition depending on the kind and source of the raw material. In Finland, for example, yields are higher in the northern part of the country, and there is a marked difference in the resin and fatty acid contents as compared with tall oil from the southern part (see Table 3) (132). American tall oils also vary considerably in composition as shown in Table 4.

American tall oil has a more resinous and less fishy odor than that produced in Finland and Sweden. Moreover, it is more difficult to distill than the Scandinavian crude oils and the resin acids do not separate out as completely (161). Tall oil from northern Finland has a higher concentration of unsaturated C_{18} fatty acids than North American tall oil (132).

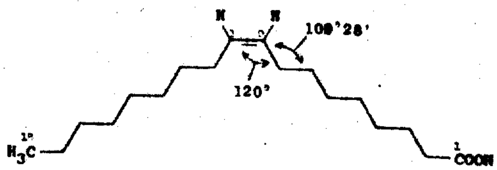
Table 3. Variation in Composition of Northern and Southern Finnish Tall Oils (132).

	Northern Finland	Southern Finland
Saturated fatty acids	6.3-6.8%	8.3-18.3%
Oleic acid	28-37%	32-43%
Linoleic acid	55-64%	38-57%
Linolenic acid	0.9-2.0%	0.5-1.0%

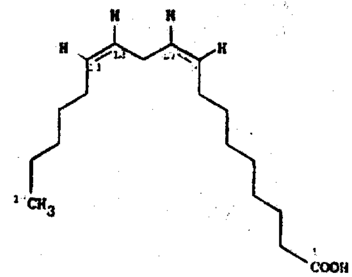
Table 4. Variation in Composition of Some U.S.A. Crude Tall Oils (161)

Tall oil	Fatty acids %	Rosin acids %	Sterols %
A	54	38	8.4
B	52	39	8.5
C	54	40	7.2
D	54	42	6.7
E	39	42	18.1
F	50	44	7.5
G	47	45	7.5
H	46	47	7.8
I	43	51	8.6
J	37	56	7.0
K	36	58	6.5

III. Structural Formulae

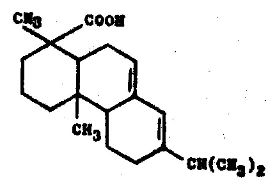


Oleic acid



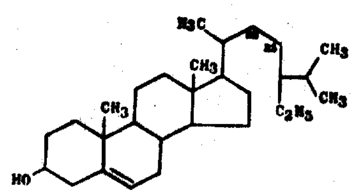
Linoleic acid

Major Tall Oil Fatty Acids



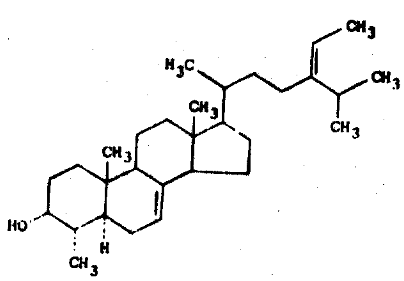
Abietic acid

Main resin acid in tall oil

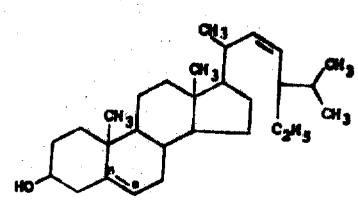


-sitosterol

Main sterol in tall oil



-sitosterol



Stigmasterol

Other sterols found in tall oil

IV. Molecular Weight - main tall oil constituents

Fatty acids:	Oleic acid	-	284.45
	Linoleic acid	-	280.44
Resin acids:	Abietic acid	-	302.44
Unsaponifiable materials:	α-sitosterol	-	426.70
	β-sitosterol	-	414.69
	Stigmasterol	-	412.67

V. Specifications

The Merck Index (139) gives the following general specifications for crude tall oil:

Dark brown liquid
Acrid odor similar to that of burnt rosin
d 0.95-1.0
n_d^{20} approx. 1.5
Acid no. 170-180
Saponification no. 172-185
Iodine no. 120-188
Fatty acids 50-60%
Rosin acids 34-40%
Unsaponifiable matter 5-10%

Specifications for tall oil rosin, tall oil pitch, and some other tall oil products are given in Table 5.

A pharmaceutical-grade tall oil sterol preparation with the following specifications has been developed for lowering the blood cholesterol (140):

Chloroform-insolubles	0.1 max.	Heavy metals	20 ppm max.
Dirt	10 ppm	Sodium	0.1% max.
Color (opt. den. at 400 mu)	0.100 max.	Methanol	0.0 ppm
Melting point	134-140°C	Total sterols[a]	95% min.
Specific rotation	-25 to -38	Unsaturated sterols	85% min.
Moisture & volatiles	2% max.	Taste	Bland
Ash	1% max.	Particle specification	1-in. diam. max.

Glycerol esters of tall oil rosin for use in chewing gum base must meet the following specifications (009):

Acid No.	5-12
Color	N or paler
Softening point	80-88°C
Ester purified by steam stripping	

[a] By digitonin assay.

Table 5. Analysis Values of Typical U.S. Tall Oil Products (064)

Product	Acid No.	Sap. No.	Rosin acids	Unsap.	Fatty acids	Color Gardner	Specific gravity 25°/25°C	Refractive index 25%
Crude tall oil	--	--	%	%	%	--	--	--
Low rosin	170	174	40.0	6.9	52.5	17	0.960	1.5030
High rosin	166	172	49.0	7.1	43.1	18	0.975	1.5100
Distilled tall oil	190	194	29.0	1.2	69.8	7+	0.947	1.4860
Acid refined tall oil	169	173	40.0	6.8	53.1	8	0.980	1.5025
Tall oil fatty acids	192	194	4.0	4.0	92.0	7+	0.901	1.4710
	196	197	1.3	1.5	97.2	5+	0.900	1.4670
	198	199	0.4	0.4	99.2	3+	0.897	1.4645
Tall oil heads	172	177	0.5	15	84.4	12+	0.905	1.4754
Tall oil rosin	168	176	94[a]	4.0	2.0	WW[b]	1.069	--
Tall oil pitch	55	115	32	32	30	17[c]	1.005	--

[a] Includes rosin esters and anhydrides. [b] U.S. rosin color standards. [c] 10% solution in benzene

Two tall oil rosin derivatives used in the manufacture of food packaging containers have the following specifications (011):

Glyceryl ester of maleic anhydride-modified tall oil rosin

Acid No.	30-40
Saponification No.	<280
Drop softening point	141-146°C
Color	N or paler

Glyceryl ester of disproportionated tall oil rosin

Acid No.	5 to 10
Saponification No.	<180
Drop softening point	84-93°C
Color	WG or paler

Oleic acid derived from tall oil fatty acids may be safely used in foods and as a component in the manufacture of food-grade additives, according to the Code of Federal Regulations, if it meets the following specifications (011):

A. The additive consists of purified oleic acid separated from refined tall oil fatty acids.

B. The additive meets the following specifications:

   1. Specifications prescribed in "Food Chemicals Codex" for oleic acid, except that titer (solidification point) shall not exceed 13.5 C and unsaponifiable matter shall not exceed 0.5%.

   2. The resin acid content does not exceed 0.01% as determined by ASTM Method D 1240-54 (1961).

   3. The requirements for absence of chick-edema factor as prescribed 121.1070.

C. It is used or intended for use as follows:

   1. In foods as a lubricant, binder, and defoaming agent in accordance with good manufacturing practice.

   2. As a component in manufacture of other food-grade additives.

The specifications for a number of American commercial tall oils and tall oil products are given in Appendix A.

VI. Description

Whole or crude tall oil is a pale amber to dark brown oily liquid which varies in consistency from a relatively thin liquid to a semi-solid "magma" of oil and crystals. It has a slightly resinous, slightly fishy odor. Physical properties and chemical composition vary considerably according to the source of the wood used and certain processing procedures. Tall oil is combustible. The flash point is 360°C.

Refined tall oil is usually produced by fractional steam distillation of crude tall oil at about 250°C, under reduced pressure of approximately 1 mm Hg. The clear amber distillate is cooled to crystallize the resin acids which are removed from the fatty acid supernatant fraction by centrifugation. High quality tall oil fatty acid preparations more than 99% pure have been produced. Resin acid fractions run as high as 97% pure.

Other tall oil raw materials and products are:

Sulfate soap - Skimmings from the spent digesting liquor in the sulfate (kraft) wood pulp process; these consist chiefly of sodium salts of fatty acids and resin acids.

Tall oil - the product formed on acidification of the sulfate soap.

Tall light oil - the first light oil resulting from the vacuum distillation of tall oil. It is composed chiefly of unsaponifiable matter but contains some entrained fatty acids.

Tall fatty acid - the entire vacuum distillate from tall oil which consists mainly of fatty acids. The highly refined grades contain more than 99% fatty acids and less than 0.5% resin acids. Ordinary grades contain from 10-20% resin acids.

Tall rosin acids - the solid crystalline substance which separates out from the distillate of tall oil and is obtained by centrifugation[a]. Its main constituent is abietic acid.

Tall rosin oil - a resinous oily material prepared by the dry distillation of tall rosin acids or tall rosin.

Tall pitch - the residue that remains after the vacuum distillation of tall oil.

Tall soap - the sodium salts of tall fatty acids.

Tall soft soap - the potassium salts of tall fatty acids.

Tall rosin soap is also prepared.

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[a] These organic acids respond to the Liebermann-Storch test for rosin and are not esterified by the Wolff procedure.

VII. Analytical Methods

The composition of tall oil, a variable mixture of fatty acids, resin acids and unsaponifiable matter, is such that it is impossible to analyze a food substance to determine if tall oil has been added. Since no constituent has yet been identified as being unique to tall oil, finding any constituent of tall oil in a substance is not conclusive evidence that tall oil has been added or used in processing.

VIII. Occurrence and Levels

Tall oil is a by-product of the wood pulp industry and does not occur naturally. The highly alkaline chemicals used in the kraft or sulfate process for digesting the lignin which binds the cellulose fibers together convert the fats and resins to soaps that are later skimmed from the spent liquor. Tall oil is obtained when the soaps (skimmings) are acidified. As mentioned earlier, it is a mixture of fatty acids, resin acids, and unsaponifiable matter.

A. Plants. Various species of pine trees are apparently the richest source of the resins and other constituents of tall oil. Since pine wood is the kind most widely used in paper making, most of the tall oil is obtained from this source. The yield of tall oil varies from 60 to 110 pounds per ton (2000 pounds) of pine pulp (90% dry) (161). Sulfate soap, the starting material for tall oil production, occurs to the extent of about 60% in the skimmings from the spent pulping liquor (Black liquor). The percentage of tall oil recovered is as high as 97% using the best modern methods. (132). One ton of crude tall oil yields approximately 500 pounds of fatty acids, 800 pounds of resin acids, and 700 pounds of secondary products (distilled tall oil, tall oil pitch, etc.).

B. Animals. Tall oil does not occur naturally in animals.

C. Synthetics. Tall oil has many commercial applications in addition to the use of certain of its refined derivatives in foods and confections and in the synthesis of certain medicinal products (cortisone, anti-cholesterol preparations, sex hormones). The ever-growing list includes: emulsifiers, anti-foam agents (yeast, antibiotic production, etc.), animal feeds, paper sizes, waterproofing agents (paper, etc.), cellulose industry, oil cloth and linoleum, coatings, rubber chemicals, synthetic rubber, wetting agents, adhesives, binders, resins, driers, soaps and other cleaners, disinfectants, fungicides, insecticides, greases, polishes, soluble oils, textile oils, gloss oils, drying oils, sulfonated oils, penetrating oils, cutting oils, boring oils, core oils, factices, flotation agents, paint, varnishes, pigment wetting agents, and printing inks (006,161).

D. Natural Inorganic Sources. There are no natural inorganic sources of tall oil.

BIOLOGICAL DATA

I. Acute Toxicity

A. Fish

As part of a study on the effect of kraft pulp mill wastes on aquatic organisms, Van Horn (152) immersed minnows, one to five per group, in varying concentrations of several tall oil products (See Table 6) in two-liter amounts at 18°C in open battery jars. The fish were observed hourly during five days for signs of toxicity. Survival times were recorded for all fatalities. In addition, determinations of pH, total alkalinity, and dissolved oxygen content were made at the conclusion of each test. If the dissolved oxygen level fell below 4 ppm, or if either of the other conditions was unfavorable for fish life, the data were discarded. Moreover, 100% survival of the untreated controls was required in each experiment. The results are presented in Acute Toxicity Table 6.

The toxicity of the tall oil resin fraction for the minnow (1 ppm) approaches that of methyl mercaptan (0.5 ppm), a better known toxic substance also found in pulp mill effluents. Crude sulfate soap and its fatty acid fraction are relatively less toxic (5 ppm). The minimum lethal concentrations of sodium oleate (5 ppm), sodium abietate (3 ppm), and phytosterol (3 ppm), all of which are compounds or derivatives normally found in tall oil, were determined also in this study.

B. Mice

The oral LD_{50} value of pale tall oil rosin in mice (strain, number, age, sex not given) following administration of the substance as a 30% solution in corn oil is given in Table 6 (177).

C. Rats

1. Weil *et al*. (136,158) administered several commercial type, epoxidized tall oil products to Carworth-Wistar male rats (4 to 5 weeks of age, 90-120 grams BW), five animals per group, in single graded doses of a logarithmic series differing by a factor of two. Water, corn oil, or semi-solid agar served as the diluent or vehicle where necessary. The animals were observed over a period of 14 days and the LD_{50} calculated from the mortality data. The results are presented in Table 6. Neither tall oil itself, nor any of its unmodified components, were included in this study since the author's primary purpose was investigation of the toxicity and possible carcinogenicity of epoxides. Data on the subject compounds are included in Table 6 because epoxidized tall oil products are sometimes used in coatings for containers that might come into contact with foods.

Table 6. Acute Toxicity of Some Tall Oil Products

	Animal	Sex & No.	Route	Dosage	Measurement	Reference
Crude sulfate soap	Fish	1-5	Immersion	5.0[a]	MLC[c]	Van Horn (152)
Sodium salts of fatty acid Fraction of crude soap	Fish	1-5	Immersion	5.0[a]	MLC[c]	Van Horn (152)
Sodium salts of resin acid fraction of crude soap	Fish	1-5	Immersion	1.0[a]	MLC[c]	Van Horn (152)
Pale tall oil rosin	Mice	---	p.o.	4600[b]	LD_{50}	Hercules (177)
Pale tall oil rosin	Rats	---	p.o.	7600[b]	LD_{50}	Hercules (177)
Epoxidized 2-ethylhexyl ester of tall oil fatty acids	Rats	5 males	p.o.	22,600[b]	LD_{50}	Weil (158)
Alkyl epoxytallate	Rats	5 males	p.o.	45,300[b]	LD_{50}	Weil (158)
Epoxidized Carbowax 200 ester of tall oil fatty acids	Rats	5 males	p.o.	39,400[b]	LD_{50}	Weil (158)
Epoxidized ethylene glycol ester of tall oil fatty acids	Rats	5 males	p.o.	53,800[b]	LD_{50}	Weil (158)
Calcium salt of epoxidized tall oil fatty acids	Rats	5 males	p.o.	20,000[b]	LD_{50}	Weil (158)
Magnesium salt of epoxidized tall oil fatty acids	Rats	5 males	p.o.	10,000[b]	LD_{50}	Weil (158)
3,4-epoxy-6-methyl-cyclohexylmethyl ester of tall oil fatty acids	Rats	5 males	p.o.	26,000[b]	LD_{50}	Weil (158)
Pale tall oil rosin	Guinea pigs	---	p.o.	4600 [b]	LD_{50}	Hercules (177)

Table 6. (Cont'd)

Substance	Animal	Sex & No.	Route	Dosage	Measurement	Reference
Epoxidized 2-ethylhexyl ester of tall oil fatty acids	Rabbits	4 males	Skin	$>$ 20,000	LD_{50}	Weil (158)
Alkyl epoxytallate	Rabbits	4 males	Skin	15,900	LD_{50}	Weil (158)
Epoxidized Carbowax 200 ester of tall oil fatty acids	Rabbits	4 males	Skin	$>$ 20,000	LD_{50}	Weil (158)

[a] Parts per million (ppm)

[b] mg/kg

[c] Lowest concentration which killed any of the fish within 5 days.

2. The oral LD_{50} of pale tall oil rosin in rats (strain, number, sex, age, weight not specified) was determined (177) using a 30% solution of the material in corn oil. The results are given in Table 6.

D. Guinea pigs

The oral LD_{50} of pale tall oil rosin in the guinea pig was determined (177). The substance was administered to the animals (strain, number, age, sex, weight not given) as a 30% solution in corn oil. The results are presented in Table 6.

E. Rabbits

Weil et al. (136,158) ascertained the topical LD_{50} values of epoxidized tall oil products in the rabbit. Albino New Zealand rabbits, 2.5-3.5 kg males in groups of four, were employed. After removing the fur from the entire trunk by clipping, the animals were immobilized and graded doses of the compounds were applied to the skin and secured for a period of 24 hours, by means of an impervious plastic covering. The film was then removed and the animals caged for a 14-day observation period. LD_{50} values were then calculated from the fatality data. Dosages greater than 20 ml/kg were not used because it was impossible to retain the material in contact with the skin for the necessary period of time. The results are presented in Table 6.

II. Short-term Studies

A. Shellfish

In a study of the effect of pulp mill wastes on York River (Virginia) oysters, Chipman et al. (030) exposed normal adult oysters to varying concentrations of the following tall oil products: strong kraft soap, weak soap, and black liquor. The effect on the pumping rate of water through the gills, after an exposure period of 30 minutes, was determined by timing the movement of a cone of carmine particles from a reservoir connected to the cloaca of the oyster (Carmine cone method). The rate of pumping before addition of the tall oil product was taken as 100% in the calculations. Untreated control oysters were included in each test. Strong kraft soap in dilutions as high as 1:200,000 had a fairly strong depressant effect on the pumping rate. Weak soap and black liquor were less toxic. Further studies showed that the toxic factor could be destroyed by oxidation; its chemical nature was not determined but the author suggested a dioxystilbene, present in pine wood, as a possibility. The effect of some pulp mill effluents on oysters is shown in Tables 7 and 8.

Table 7. Mean Values of Hours Open in Various Concentration of Pulp-Mill Effluent (030)

Concentration in Parts per 10 Liters	Mean Value of Hours Open	Standard Deviation	Standard Error
1,000	5	...	...
100	8.5	...	...
50	12.8	2.40	± 0.85
25	13.7	2.52	± 0.89
10	13.5	3.89	± 1.17
5	17.9	1.29	± 0.65
2.5	20.5	3.15	± 1.57
2.0	19.2	1.63	± 0.67
1.0	20.9	2.06	± 1.03
0	21.3	1.29	± 0.26

Table 8. Depression of the Rate of Filtration Caused by Pulp-Mill Effluent (030)

[Specific gravity 1.0043. Carmine-cone method. Figures express means of percentage of normal rate. Each oyster was exposed to one concentration of the effluent for 1-1/2 hours. Three sets of readings were made at half-hours intervals.]

Concentration, parts per liter	Number of oysters	Percentage of Normal Rate	Standard Deviation	Percentage of Depression
0	21	100	....	0
0.25	6	99.2	4.37 ± 0.87	0.8
0.5	18	89.2	8.21 ± 1.90	10.8
1.0	27	77.8	5.83 ± 1.12	22.2
2.0	15	36.5	5.65 ± 1.46	63.5

B. Fish

1. Nehring (105) exposed two species of fish - perch (Perca fluvialis) and roaches (Rutilus rutilus), average length, 13-18 cm, to varying concentrations of tall oil in water (10 liters) in well-ventilated glass aquaria at a temperature range of 11-17°C for a period of 3-4 days. The tall oil used had the following specifications: fatty acids (40-55%), resin acids (30-40%), unsaponifiable matter (12.5%), petroleum ether-insoluble residue (2%). Ethanol (amount not given) was added to the tall oil to promote emulsification with the aquarium water. Apparently the amount used was not toxic.

C. Chicks

1. Sunde (142) fed chicks tall oil and tall oil resin acids as part of a general study on the value of fats as supplements for chick rations. The crude tall oil used (Ligro[a]) consisted of 46-52% fatty acids, and 41-52% resin acids. The refined oil was a distillate from the crude oil consisting of 61-71% fatty acids (oleic acid, 45, linoleic acid, 48, palmitic acid, 6-7) and 25-30% resin acids. The tall oil rosin acids fraction, crystallized from crude tall oil, contained about 90% resin acids (abietic acid 30-40, neoabietic acid, 10-20, and other resin acids in smaller amounts).

Day-old chicks of both sexes, progeny of New Hampshire males and Single Comb White Leghorn females, 25 chicks per group, were fed the tall oil products at levels of 5% in Basal diet A for a period of 4 weeks. Tenox II[b] was added in the amount of 0.5% to all fatty acids used in the study except stearic acid and the hydrogenated fats. Control groups on the basal diet alone were included in each experiment. Effects of the supplements on growth rate, feed utilization (grams feed/g weight), and percent fat in the feces were determined; results are presented in Table 10.

All of the tall oil products had a depressing effect on growth. The rosin acids fraction had the most severe effect; refined tall oil was the least deleterious; crude tall oil (Ligro) was intermediate between the other two in its effect. Sunde concluded that the growth-depressing effect of refined tall oil was due to resin acids not removed by distillation (25-30%).

Feed utilization (grams feed/gram weight) was also reduced by refined as well as crude oil. This determination was not made with the rosin-fed chicks.

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[a] Manufactured by West Virginia Pulp & Paper Company, Charleston, South Carolina.

[b] Butylated hydroxyanisole (20%), propyl gallate (6%), Citric Acid (4%), and Propylene glycol (70%)

Table 9. Toxicity of Tall Oil for Fish (105)

Substance	Animal	Age & Wt.	Route	Dosage	Measurement	Reference
Tall Oil	Fish (Perch)	13-18 cm in length	Immersion	10-20[a]	Threshold[b] value	Nehring (105)
Tall Oil	Fish (Roach)	13-18 cm in length	Immersion	20-40[a]	Threshold[b] value	Nehring (105)

[a] mg/liter

[b] Concentration of the substance at which the first clearly-detectable poisoning symptoms appear in an aquarium test within 3 to 4 days

Table 10. The Effect on Chicks of Tall Oil Products, and Fatty Acids on Body Weight, Feed Conversion and Feces Fat (142)

Experiment 8

	Wt.(gms)	Gms. feed / Gm. wt.	% fat in dry feces
Basal diet A	302.2	2.04	1.12
	312.5	2.05	1.20
Basal + 5% crude tall oil	155.8	3.05	--
Basal + 5% tall oil	201.7	3.01	2.44
Basal + 5% rosin acids	88.9	--	--
Basal + 5% oleic acid	282.7	1.95	1.89
Basal + 5% butyric acid	204.7	1.95	.94
Basal + 5% linolenic acid	327.3	1.89	2.14
Basal + 5% white grease	290.1	1.92	1.27
Basal + 5% linoleic acid	316.3	1.91	1.45

2. Antila *et al*. (014) investigated the effect of tall oil fatty acid ethyl esters as a dietary supplement on egg production, fertility, hatchability, and composition of carcass lipids in a series of three experiments lasting from 3 to 6 months. In each experiment, White Leghorn hens, 23 to 63 per group, were fed tall oil fatty acid ethyl esters at levels of 5% and 10% in a basic diet. Food and water were allowed *ad libitum*. In addition, a grain mixture (one-half oats and one-half barley) was provided in the amount of 60 grams per hen per day. A control group, fed the basic diet and grain mixture only, was included in each experiment. Food consumption and egg production were determined for each group. Egg weight, fertility, hatchability, and certain other characteristics were determined on eggs selected at random from each group. Effect of the compounds on carcass fat composition was ascertained by fatty acid analysis. The results are given in Tables 11-17.

Although no fatalities attributable to the tall oil fatty acid ethyl esters were mentioned, the higher level (10% in diet) definitely reduced food consumption in all experiments. The authors attributed this to appetite impairment which could, of course, be a manifestation of toxicity. At the lower fat level (5% in diet), food consumption was lower than that of the controls in the first experiment, but not in the second and third experiments. The grain ration was consumed completely by all groups in each experiment (Tables 11-13).

Table 11. Feeding Test, May to July 1962 (014)

	Month	Average number of hens	Egg production (%)	Dry feed mixture g	Grains g	Total g	Ethyl ester in the total feed ration g	Ethyl ester in the total feed ration %
Group No. 1	IV	27.0	63.5					
(Controls)	V	27.0	64.1	81	60	141		
	VI	25.5	64.4	71	60	131		
	VII	23.8	62.8	69	60	129		
Group No. 2	IV	52.0	61.0					
5 % ethyl ester	V	51.4	54.8	50	60	110	2.5	2.3
in the dry feed	VI	46.7	55.0	54	60	114	2.7	2.4
mixture	VII	45.8	59.9	62	60	122	3.1	2.6
Group No. 3	IV	64.0	59.7					
10 % ethyl ester	V	63.5	55.6	55	60	115	5.5	4.8
in the dry feed	VI	56.0	52.7	45	60	105	4.5	4.3
mixture	VII	52.9	50.6	50	60	110	5.0	4.5

Table 12. Feeding Test, December 1962 to May 1963 (014)

	Month	Average number of hens	Egg production (%)	Dry feed mixture g	Grains g	Total g	Ethyl ester in the total feed ration g	%
Group No. 1	XII	36.7	72.7	80	60	140		
(Controls)	I	36.0	68.1	53	60	113		
	II	35.4	73.1	59	60	119		
	III	34.9	68.0	47	60	107		
	IV	33.4	66.4	52	60	112		
	V	29.1	69.0	78	60	138		
Group No. 2	XII	30.0	85.2	69	60	129	3.5	2.7
5% ethyl ester	I	30.0	82.7	57	60	117	2.9	2.4
in the dry feed	II	29.4	84.4	63	60	123	3.2	2.6
mixture	III	29.0	75.4	51	60	111	2.6	2.3
	IV	29.0	76.2	82	60	142	4.1	2.9
	V	26.2	71.6	83	60	143	4.2	2.9
Group No. 3	XII	29.3	71.1	51	60	111	5.1	4.6
10% ethyl ester	I	26.8	63.3	40	60	100	4.0	4.0
in the dry feed	II	26.0	63.2	30	60	90	3.0	3.3
mixture	III	25.1	63.4	30	60	90	3.0	3.3
	IV	25.0	61.7	50	60	110	5.0	4.5
	V	23.1	60.1	47	60	107	4.7	4.4

Table 13. Feeding Test, February to April 1964 (014)

	Month	Average number of hens	Egg production (%)	Dry feed mixture g	Grains g	Total g	Ethyl ester in the total feed ration g	%
Group No. 1	II	39.0	79.6	77	60	137		
(Controls)	III	39.0	78.1	90	60	150		
	IV	38.1	68.8	69	60	129		
Group No. 2	II	42.0	81.2	60	60	120	3.0	2.5
5% ethyl ester in	III	42.0	77.5	70	60	130	3.5	2.7
the dry feed mixture	IV	41.0	74.7	64	60	124	3.2	2.6
Group No. 3	II	47.0	77.0	52	60	112	5.2	4.6
10% ethyl ester in	III	47.0	72.8	57	60	117	5.7	4.9
the dry feed mixture	IV	45.1	70.0	50	60	110	5.0	4.5

The authors interpreted the variable egg production data as indicating a detrimental effect of the tall oil preparation (Tables 11-13). Fertility and hatchability, on the other hand, were not impaired (Table 14).

Marked changes occurred in the fatty acid composition of the yolk in response to feeding the tall oil products. The amount of oleic acid increased significantly whereas the concentrations of palmitic, stearic, and linoleic acids were correspondingly lowered (Table 15). The tall oil esters also caused a reduction in egg weight and impaired baking characteristics (Table 16).

Table 14. Fertilization and Hatching Percentage of Eggs (014)

Group	Fertilization percentage			Hatching percentage		
		Ethyl ester in the dry feed mixture			Ethyl ester in the dry feed mixture	
	Controls	5 %	10 %	Controls	5 %	10 %
First hatching	94.6	87.9	89.2	80.0	81.3	76.9
Second hatching	93.2	87.7	90.8	83.8	79.2	79.2
Mean	93.8	87.8	90.0	81.9	80.2	78.0

In order to clarify the composition and properties of the eggs, the weights of shell, egg white and yolk were separately determined for 15 random samples from each feeding group. Furthermore, the fat content of the yolk was determined and the iodine number and fatty acid composition of the isolated lipids were determined. In conditions con-

Table 15. Effect of Ethyl Ester Feeding on the Fatty Acid Composition of the Eggs Lipids (014)

Ethyl ester in the dry feed (%)	Feeding test No.	Fat content %	Iodine number of the fat	Fatty acid composition (%)					
				C_{14}	C_{16}	$C_{16}1$ =	C_{18}	$C_{18}1$ =	$C_{18}2$
0	2	31.7	77.3	0.8	29.6	2.8	13.9	35.5	17.1
	3		77.5						
5	2	32.5	77.4	0.6	26.3	4.1	10.7	43.4	15.0
	3		78.8						
10	2	32.7	78.4	0.6	25.2	4.8	8.5	47.8	13.3
	3								

Table 16. Effect of the Feeding of Ethyl Esters of the Fatty Acids of Tall Oil an the Weight of the Eggs (014)

The figures refer to 15 eggs each.

Ethyl ester in the dry feed (%)	Feeding test No.	Weight of the eggs, g	Shell weight		Egg-white weight		Yolk weight		Handling losses per cent units
			g	%	g	%	g	%	
0	2	897.7	86.2	9.6	533.4	59.4	266.9	29.7	1.3
	3	914.0	85.3	9.3	537.8	58.8	276.4	30.2	1.7
5	2	889.6	85.2	9.6	521.6	58.6	266.9	30.0	1.8
	3	885.3	89.0	10.5	512.3	58.1	267.4	30.5	0.9
10	2	861.0	85.4	9.9	504.3	58.6	263.7	30.6	0.9
	3	872.7	89.8	10.3	506.9	58.1	268.8	30.8	0.8

In contrast to the effect on yolk lipids, tall oil fatty acid ethyl esters caused no alternation in the composition of carcass fat in the chick (See Table 17).

Firestone et al. (048) recently discovered the chick-edema factor in tall oil. This substance, found in toxic fats, has caused large numbers of chick fatalities. A food additive regulation of the FDA now requires that food grade fatty acids be free of the chick-edema or other toxic factors (Code of Federal Regulations, Title 21, Section 121.1070) (011). The exact chemical nature of this factor has not been determined as yet, but considerable evidence points toward the chlorinated aromatic hydrocarbons (048).

D. Rats

1. Young albino rats (strain, age, sex, weight, and numbers not specified), fed pale tall oil, gum, and wood rosin in the diet at levels of 5, 1, 0.2, 0.05, and 0.01 percent for 90 days, were observed for general appearance, food consumption, and growth (177). Hematologic and urine analyses were performed also. At the conclusion of the 90-day period, all surviving animals were sacrificed and examined for gross and microscopic pathologic changes in heart, liver, kidneys, spleen, brain, and a number of other organs.

At the 5% level, all of the rats died within two weeks. Death was preceded by marked food refusal and a rapid weight loss.

At the 1% level, no fatalities occurred although food intake and growth were below normal at first. After two weeks, however, both rates were comparable to those of the untreated controls. Urine and hematologic values were normal throughout. At autopsy the test animals were found to have higher liver weights than the controls, but other organ weights were normal. No significant histopathologic alterations were seen in any of the organs, including the liver.

At the 0.2, 0.05, and 0.01% levels, no significant differences were noted between treated and control animals.

Table 17. Effect of Ethyl Ester Feeding in the Fatty Acid Composition of Hens' Carcass Fat (014)

	Ethyl ester administered at		
	0%	5%	10%
Iodine number of the fat . . .	84.2	83.3	85.6
C_{14}	0.4	0.4	0.5
C_{16}	19.1	20.6	16.9
$C_{16}1=$	2.9	3.3	2.7
C_{18}	5.8	5.5	5.1
$C_{18}1=$	46.9	47.8	46.0
$C_{18}2=$	24.8	21.1	26.5
$C_{18}3=$	0.7	0.5	0.9
C_{20}	1.0	0.7	0.8

2. Weil et al. (136,158) subjected albino male or female rats (strain, age, weight not specified), in groups of six, to the concentrated vapor of several epoxidized tall oil products for periods ranging from one-fourth to eight hours, in a logarithmic series with a ratio of two. They then determined the longest inhalation period which permitted all of the animals to survive during the two-week subsequent observation period.

All of the preparations were relatively non-toxic, permitting 100% survival at the maximum inhalation period of eight hours. Epoxidized soybean oil, glycidyl oleate, and all other edible lipoidal substances included in the test were likewise relatively non-toxic. In contrast, however, propylene oxide killed all of the test animals within five minutes and the maximum survival time with glycidyl acrylate was only thirty minutes.

3. Seppanon and coworkers (131,132), in a series of thorough and detailed experiments, investigated the effect of a number of tall oil fatty acid products as dietary supplements in the nutrition of rats. These studies are prompted in the main by discoveries that indicated the possibility of preparing suitable food fats from tall oil. For example, the ethyl and glyceryl esters of refined tall oil fatty acids resemble edible oils like soybean oil in many respects.

In the preliminary experiment, Seppanen et al. (131) fed weanling Sprague-Dawley male or female rats (40-50 g BW, ten per group) various tall oil preparations at levels of 13.5% and 35% (30% and 60% of the total calories) in a basal diet[a] for periods of three weeks (one 2-1/2 month experiment was conducted also). Control groups received corresponding amounts of soybean oil, butter, or commercial margarine. The animals were kept in individual wire-bottom cages; food and water were made available ad libitum. In the short-term experiments, effect on growth was ascertained on the basis of growth curves plotted from average body weights at various intervals. In the "long-term" study, effects on reproduction and on several internal organs were determined also. The results are presented in Figures 3-7.

Both refined and unrefined glyceryl, and ethyl esters of tall oil fatty acids had a marked inhibitory effect on the growth of rats when fed at levels of 13.5% and 35% in the diet. The effect of the unrefined ethyl product was especially pronounced causing death of the entire group within ten days (see Fig. 3).

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[a] Graham flour, casein, dried brewer's yeast, and salt mixture.

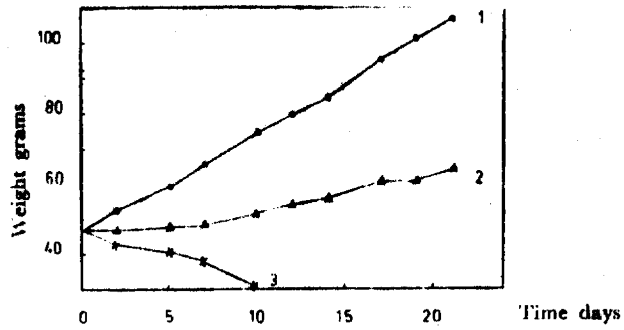

Figure 3. Average body Weights of Rats Fed 30% of the Calories from Ethyl Ester of Soybean Oil (1), Refined Esters of Tall Oil Fatty Acids (2) or Unrefined Ethyl Esters of Tall Oil Fatty Acids (3).

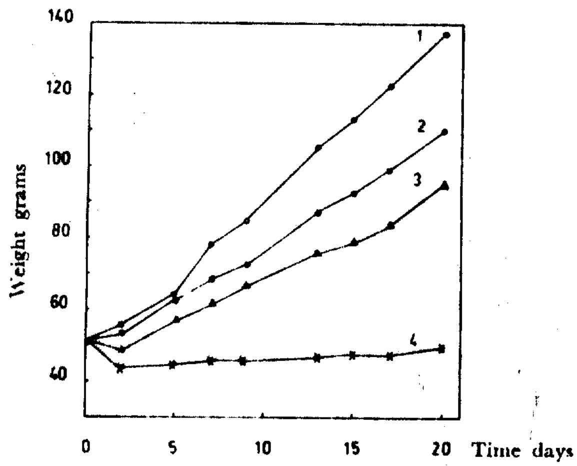

Figure 4. Average Body Weights of Rats Fed 30 or 60% of the Calories From Soybean Oil (1 and 2) and 30 or 60% of the Calories from Glyceryl Esters of Tall Oil Fatty Acids (3 and 4)

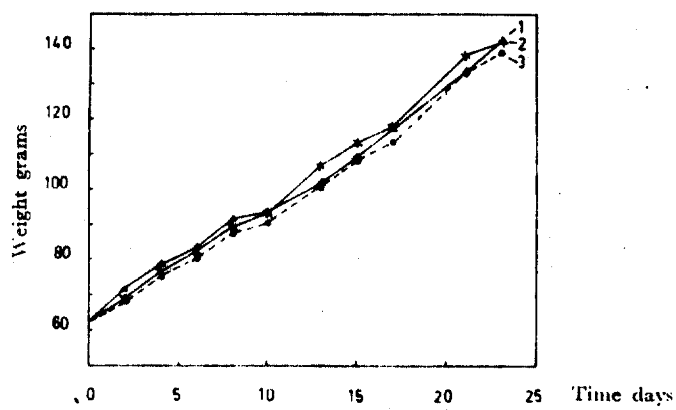

Figure 5. Average Body Weights of Rats Fed 30% of the Calories from Hydrogenated Soybean Oil (1), Soybean Oil (2) or Hydrogentated Glyceryl Esters of Tall Oil Fatty Acids (3).

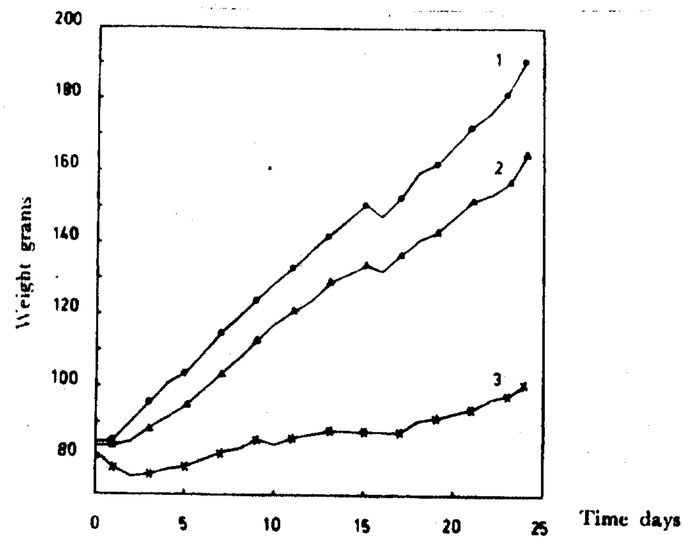

Figure 6. Average Body Weights of Rats Fed 30% of the Calories From Ethyl Ester of Linseed Oil (1), Ethyl Esters of Tall Oil Fatty Acids(2) or Ethyl Ester of cis-5,9,12-Octadecatrienoic Acid (3).

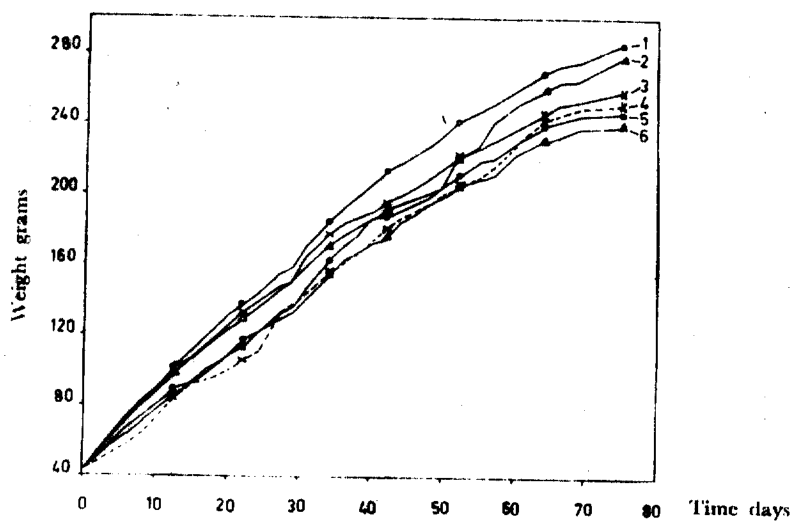

Figure 7. Average Body Weights of Male Rats Fed 30% of the Calories from Butter (1), Tall Oil Margarine (2) or Margarine (3) and 60% of the Calories from Margarine (4), Butter (5) or Tall Oil Margarine (6).

Hydrogenation experiments indicated that the growth-retarding principle was probably associated with unsaturated fatty acids since rats fed on hydrogenated glycerylesters tall oil product grew practically as well as the hydrogenated or unhydrogenated soybean oil controls (see Fig. 5). Further experiments seemed to incriminate cis-5,9,12-octadecatrienoic acid in particular (see Fig. 6). The matter is complicated, however, by the results of another experiment where this acid from a different source did not retard growth (131).

A "margarine - type" tall oil fatty acid product, prepared by interesterification of hydrogenated refined tall oil fatty acid glycerides and hydrogenated soybean oil, compared very favorably with butter and ordinary margarine in growth-promoting properties (see Fig.7). Reproduction was not adversely affected, and no histopathologic changes were detected in the heart, liver, kidneys or thyroid gland.

Seppanen (132) continued investigation of tall oil products as dietary supplements in a more extensive and detailed series of experiments on the growth of rats published in 1969. The fatty acids, resin acids, and unsaponifiable fractions were studied as well as the refined esters (ethyl and glyceryl) and hydrogenated interesterified tall oil margarine. Various fatty acid sub-fractions and cis-5,9,12-octadecatrienoic acid concentrate were investigated also in an effort to determine the nature of the growth-retarding factor(s). The mortality data for this series of experiments are summarized in Table 18.

Male and female, Sprague-Dawley rats were used throughout. The basal diet was composed of graham flour, casein, dried brewer's yeast, salt, and vitamin mixtures. The tall oil products or control fats (soybean oil, butter or margarine) were added to the basal diet at the following levels:

Diet Type	Fat or Oil %wt.	Fat or Oil % of total calories	Fat-soluble vitamin supplement
A	6.1	15	Synthetic
B	13.1	30	Synthetic
C	34.6	60	Synthetic
D	13.2	30	Cod liver oil
E	34.8	60	Cod liver oil

The exact compositions are given in the referenced paper.

Table 18. Tall Oil Fatty Acid Products in the Diet of Rats - Mortality Data (132)

Product	Percent in diet	Mortality
Tall oil fatty acids distillate	6.1	1/10
Tall oil fatty acids distillate	13.1	0/10
Tall oil fatty acids distillate	34.6	10/10[a]
Ethyl esters of tall oil fatty acids	13.2	0/10
Ethyl esters of tall oil fatty acids	34.6	12/18
Glyceryl esters of tall oil fatty acids	13.2	1/20
Glyceryl esters of tall oil fatty acids	34.6	13/26
Hydrogenated glyceryl esters of tall oil fatty acids	13.2	0/10
Tall oil fatty acids glyceride margarine	13.2	2/34[b]
Tall oil fatty acids glyceride margarine	34.8	0/15[b]
Soybean oil control	6.1	0/10
Soybean oil control	13.2	2/30
Soybean oil control	34.8	2/46
Butter control	13.2	1/34[b]
Butter control	34.8	0/15[b]
Commercial margarine control	13.2	3/34[b]
Commercial margarine control	34.8	3/15[b]

[a] All animals died within 4 days

[b] Experiment I only

The rats were caged individually and food and water given _ad libitum_. Food consumption and growth rates were determined.

The short-term experiments were 2-4 weeks in length; male rats only were used. The "long-term" studies lasted two to three times longer and both male and female rats were used. The animals were observed closely throughout for toxic effects of the tall oil products as well as their influence on growth.

4. In the first experiment of this series, Seppanen (132) fed groups of ten young male rats a distillate of tall oil fatty acids at levels of 6.1, 13.1, and 34.6% (15, 30, and 60% of total calories) in diet types A, B, and C, respectively, for a period of four weeks. Corresponding control groups were given the same amount of soybean oil. Food consumption toxic effects and influence on the growth rate were determined as indicated above. The experimental design and results are shown in Table 19 and Figure 8.

At the higher level (34.6%), all of the animals on the tall oil ration died within a few days. (The only other fatality was one of ten animals at the lower test compound level). At the 13.1% level, food consumption of the tall oil test group was depressed to a little more than half that of the control group and growth was definitely retarded. The final weight difference between test and control animals was highly significant statistically ($P < 0.001$).

5. Young rats (48-49 g.), ten per group, were fed ethyl esters of distilled tall oil fatty acids by Seppanen (132) at levels of 13.1 and 34.6% in diet types B and C for periods of ten days and two weeks, respectively. Control groups were fed soybean oil. The results are given in Table 20.

At the higher level of the test product, six of ten young rats died; in the adult group, only one animal succumbed. Skin lesions were observed, however, and the animals had glossless shaggy fur. All control animals were normal. No deaths occurred in any of the other groups.

Food consumption and growth rates were depressed at both test levels in the adult and young groups. At the higher level of the tall oil preparation, the difference between test and control groups was highly significant statistically ($P < 0.0001$); at the lower level, the difference was significant ($P < 0.01$).

6. Glyceryl esters of distilled tall oil fatty acids, the tall oil products which resemble edible vegetable oils most closely, were studied by Seppanen (132).

Table 19. **Effect of Tall Oil Fatty Acid Distillate on the Weight Gain of Young Rats (132)**

Type and amount of fat in the diet		Mean initial weight, g	Average cumulative weight gain, g — Week of experiment 1.	2.	3.	4.	Deaths	Average food consumption, g
Tall oil fatty acid distillate	15 cal %	40.3	16.2	39.4	63.6	95.5	1/10	274.7
Tall oil fatty acid distillate	30 »	40.1	4.6	16.6	35.9	55.3	0/10	170.5
Tall oil fatty acid distillate	60 »	39.9	—[1]	...	—	—	10/10	—
Soybean oil	15 »	39.8	23.0	50.5	80.3	101.5	0/10	316.3
Soybean oil	30 »	40.0	30.0	59.6	98.9	128.1	0/10	313.6
Soybean oil	60 »	40.1	17.9	35.3	57.6	85.1	0/10	205.6

[1] All animals died within 4 days.

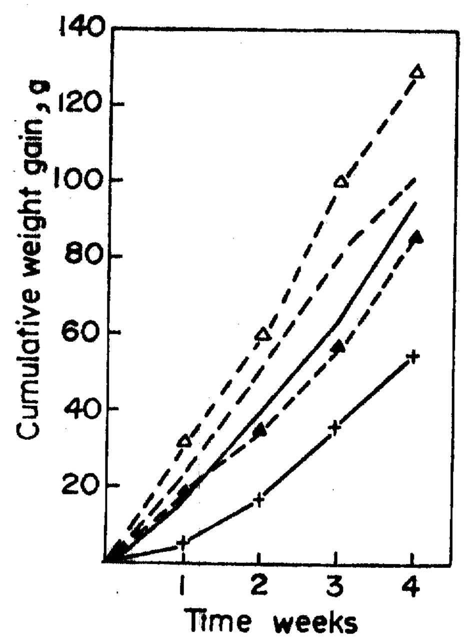

Figure 8. **Cumulative Growth Curves for Rats Fed Tall Oil Fatty Acid Distillate and Soybean Oil Fatty Acids. (132)**

———— 15 cal % tall oil fatty acid distillate , **6.1% by weight**
—×—×—×— 30 » » » » » , **13.1% by weight**
– – – – – 15 » soybean oil fatty acids , **6.1% by weight**
—△—△—△— 30 » » » » , **13.1% by weight**
—▲—▲—▲— 60 » » » » » , **34.6% by weight**

He fed young rats (51-52 g.), ten per group clarified[a], as well as the unclarified product, at levels of 13.2% and 34.8% in diet types D and E for a period of 20 days. Food consumption and growth rates were determined as before and the animals were observed for symptoms of toxicity throughout. The results are given in Table 21.

At the higher fat level, both products were highly toxic. Six animals in one group and five in the other succumbed. No deaths occurred in the corresponding control groups. At the lower fat level, two animals in one test group and one in the other died. One control animal also succumbed, however.

Food consumption and weight gain of animals on the tall oil products at both levels were considerably less than the figures for the corresponding control animals. The growth rate differences were highly significant statistically. ($P < 0.001$).

7. Since tall oil fatty acid distillate contains a small amount of resin acids (ca 2%) and unsaponifiable matter, Seppanen (132) fed young rats (40-50 g.), six per group, each of these materials (5 parts + 95 parts soybean oil) at levels of 34.8% in diet type E for a period of 21 days. The object was to determine their possible detrimental effect on growth. Several other tall oil preparations were included also, with soybean oil being used as the control. The overall design of the experiment and the results obtained are presented in Table 22.

One-third of the animals fed unsaponifiable matter and one-sixth of the rats on the resin acids diet, succumbed. No deaths occurred in the soybean oil control group. Two-thirds of the animals fed the glycerol esters of tall oil fatty acids distillate died. There were no fatalities in any of the other test groups.

Growth of all animals on the test diets was poorer than those on the soybean control diet. The difference was significant statistically ($P < 0.01$) for the resin acids and unsaponifiable matter groups. The average food consumption of these groups was also significantly below that of the control group.

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[a] Produced by cooling the ordinary glycerol esters of tall oil fatty acids distillate to 4°C. and removing the insoluble material by centrifugation.

Table 20. Effect of Ethyl Esters of Tall Oil Fatty Acid Distillate on Weight Gain of Rats (132)

Type and amount of fat in the diet		Mean initial weight, g	Average cumulative weight gain, g						Deaths	Average food consumption, g
			Day of experiment							
Young rats			2.	5.	6.	8.	10.	12.		(10 days)
Ethyl esters of tall oil fatty acids	60 cal %	48.7	−2.5	−5.0	−6.4	−7.0	−7.0	−3.2	6/10	39.0
Soybean oil	60 »	48.4	4.8	12.9	15.8	24.5	32.2	40.3	0/10	79.4
			Week of experiment							
Adult rats			1.		2.	3.		4.		(2 weeks)
Ethyl esters of tall oil fatty acids	30 »	229.2	0.8		16.7	8.4		26.9	0/10	225.2
Ethyl esters of tall oil fatty acids	60 »	229.7	−31.6		−36.4	−53.0		−42.5	1/10	107.9
Soybean oil	30 »	229.4	15.0		48.2	51.8		74.9	0/10	258.8
Soybean oil	60 »	229.8	12.1		40.9	60.9		69.8	0/10	117.8

Table 21. Effect of Glyceryl Esters of Distilled Tall Oil Fatty Acids on Weight Gain of Young Rats (132)

Type and amount of fat in diet		Mean initial weight, g	Average cumulative weight gain, g								Deaths	Average food consumption, g
			Day of experiment									
			2.	5.	7.	9.	13.	15.	17.	20.		
Glyceryl esters of tall oil fatty acids	30 cal %	51.6	−2.8	5.0	9.5	14.5	24.2	27.1	31.0	43.3	1/10	132.2
Glyceryl esters of tall oil fatty acids	60 »	51.6	−7.9	−7.1	−6.2	−6.0	−5.2	−4.5	−4.5	−1.8	5/10	61.3
Clarified glyceryl esters of tall oil fatty acids	30 »	51.7	−2.1	5.2	9.6	12.4	19.6	26.5	30.5	44.7	2/10	123.9
Clarified glyceryl esters of tall oil fatty acids	60 »	51.7	−7.5	−7.3	−6.8	−6.4	−2.5	1.1	−0.3	3.3	6/10	54.5
Soybean oil	30 »	51.5	3.7	17.9	26.7	33.4	54.1	61.9	70.8	86.1	1/10	186.8
Soybean oil	60 »	51.6	1.3	11.3	17.1	21.4	35.5	41.0	47.4	58.4	0/10	127.0

Table 22. Effect of Different Fractions of Glyceryl Esters of Distilled Tall Oil Fatty Acids on Weight Gain of Young Rats (132)

Type and amount of fat in diet		Mean initial weight, g	Average cumulative weight gain, g Day of experiment 2.	5.	7.	10.	12.	14.	19.	21.	Deaths	Average food consumption, g
Glyceryl esters of tall oil fatty acids	60 cal %	49.1	−3.6	−6.1	−7.1	−5.4	−5.1	6.9	10.9	9.9	4/6	74.5
Soybean oil/glyceryl esters of tall oil fatty acids 90:10	60 »	49.1	4.5	13.2	19.9	28.2	33.2	40.7	50.7	58.5	0/6	120.0
Soybean oil/glyceryl esters of elaidin precipitate 90:10	60 »	49.3	4.8	9.7	18.5	26.3	31.7	39.3	55.5	65.2	0/6	133.4
Soybean oil/unsaponifiable matter of tall oil fatty acid distillate 95:5	60 »	49.3	5.3	11.8	19.5	23.0	27.5	29.7	47.4	53.9	2/6	119.5
Soybean oil/resin acids of tall oil fatty acid distillate 95:5	60 »	49.3	−0.8	2.0	8.0	19.7	24.9	29.9	45.9	54.1	1/6	109.3
Soybean oil	60 »	49.3	3.5	13.5	21.8	30.5	38.7	44.9	64.5	73.9	0/6	134.1

Table 23. The Effect of the Molecular Distillate of the Glyceryl Esters of Distilled Tall Oil Fatty Acids on Weight Gain of Young Rats (132)

Type and amount of fat in the diet		Mean initial weight, g	Average cumulative weight gain, g Day of experiment 2.	4.	7.	10.	12.	14.	18.	21.	Deaths	Average food consumption, g
Molecular distillate of glyceryl esters of tall oil fatty acids	60 cal %	61.5	−3.4	−5.0	−4.9	−1.9	−2.6	−1.4	1.7	2.3	1/10	90.6
Soybean oil	60 »	61.5	3.2	6.6	13.5	24.0	28.3	37.2	48.3	63.1	0/10	156.1

Table 24. The Effect of Ethyl Esters of Fractions of Tall Oil Fatty Acid Distillate Separated with Urea on Weight Gain of Young Rats (132)

Type and amount of fat in the diet		Mean initial weight, g	Average cumulative weight gain, g Day of experiment 2.	4.	6.	8.	10.	Deaths
Ethyl esters of tall oil fatty acids	60 cal %	48.8	−4.1	−0.3	−3.8	−4.3	−5.8	6/8
Esterified tall oil fatty acid fraction 1	60 »	48.8	0.1	3.6	7.1	11.9	17.5	1/8
Esterified tall oil fatty acid fraction 2	60 »	48.8	−5.2	−3.2	−3.5	−3.8	−5.0	4/8
Esterified tall oil fatty acid fraction 3	60 »	48.8	−6.5	−5.3	−4.3	−4.8	−7.3	6/8
Esterified tall oil fatty acid fraction 4	60 »	48.6	−10.2	—[1]	—	—	—	8/8
Ethyl esters of soybean oil » »	60 »	48.6	3.3	11.8	17.7	25.7	35.4	0/8

[1] All animals died within 3 days.

8. A fraction of tall oil fatty acids distillate, freed from most of the unsaponifiable matter by molecular distillation, was fed by Seppanen (132) to a group of ten young rats (61.5 g), at a level of 34.8% in diet type E[a] for a period of 21 days. The control group received soybean oil. Food consumption, cumulative weight gain, and toxic effects were determined in the usual manner. The results are given in Table 23.

Of the ten animals in the test group, only one died during this experiment compared with fatality rates of 50% and 60% in an earlier experiment of this series when undistilled esters (molecular mtd.) were fed at the same level. In spite of the low death rate, however, the test group animals lost weight from the beginning until near the end of the experiment when there was a slight gain but not enough to reach the original level. The difference between weight gain of the control and test groups was highly significant statistically ($P < 0.001$). The average food consumption of the test group was considerably lower also than that of the control group.

9. Seppanen (132) studied the effect of four fractions of tall oil fatty acids on the growth of rats. The products were prepared by a urea fractionation procedure and then esterified (ethyl esters) before incorporation in the basal diet. The control preparations were ethyl esters of unfractionated tall oil fatty acids and ethyl esters of soybean oil.

Young rats (40-50 g), eight per group, were fed the various preparations at a level of 34.8% in diet type E for a period of ten days. Toxic effects and influence on growth are presented in Table 24.

The major discovery in this short experiment was the exceptionally high toxicity of Fraction 4 which contained most of the *cis*-5,9,12-octadecatrienoic acid; all animals in this group died within three days. Fractions 2 and 3 were very toxic also at the high level used, with fatality rates of 50% and 75%, respectively. Fraction 1, on the other hand, was considerably less toxic with only one of eight animals dying. The feeding period lasted only ten days, however, and even during this short time the cumulative weight gain was just about half that of the soybean oil control group ($P < 0.01$).

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[a] Five percent of the total fat in this particular diet consisted of soybean oil to assure an adequate amount of essential fatty acids.

10. Since the location of the double bonds in cis-5,9 12-octadecatrienoic acid are uncommon in natural lipids, it appeared that this unusual acid might be responsible, in part at least, for the growth-retarding and toxic effects of tall oil fatty acid distillate. Seppanen (132) therefore fed a group of ten young rats (81-84 g) the ethyl esters of 80% concentrate of cis-5,9,12-octadecatrienoic acid at a level of 13.2% in diet type D for a period of 25 days. Control groups of the same number and specifications were fed the ethyl esters of tall oil fatty acids or ethyl esters of linseed oil fatty acids. Average food consumption, growth rates, and toxic effects were determined in the usual manner. The results are given in Table 25.

Although no deaths occurred, food consumption and the growth rate were strongly depressed in the cis-5,9,12-octadecatrienoic acid group. Differences in weight gains between this group and the controls was highly significant statistically ($P < 0.001$).

11. In another experiment on the effect of cis-5,9,12-octadecatrienoic acid on the growth of rats, Seppanen (132) used pine seed oil as the fat ingredient in the diet. Pine seed oil contains twice as much of this unusual acid as is in the tall oil fatty acid distillate. Soybean oil extract, obtained in the same way as the pine seed oil, was used as one control fat; ordinary soybean oil was used as the other.

In this experiment, groups of ten young rats (46-47 g.) were fed the products at a level of 13.2% in the diet type D for a period of 16 days. Food consumption, growth rates, and toxic effects were determined as before. The results are presented in Table 26 and Figure 9.

No deaths occurred in the pine seed oil group. Moreover, the growth rate almost equaled that of the soybean controls (the differences were not significant statistically). Seppanen concluded on the basis of these results that cis-5,9 12-octadecatrienoic acid was not the growth-retarding principle in tall oil fatty acids distillate. He speculated that the toxic substance was more likely produced during distillation.

12. A preparation of hydrogenated glyceryl esters of tall oil fatty acids was used in this experiment. After refining and bleaching,this product closely resembles food fats in many respects including chemical and physical constants. Groups of ten young rats (62-63 g) were fed the hydrogenated

Table 25. The Effect of Ethyl Esters cis-5,9,12-octadecatrienoic Acid on Weight Gain of Young Rats. (132)

Type and amount of fat in the diet		Mean initial weight, g	Average cumulative weight gain, g — Day of experiment								Deaths	Average food consumption, g
			2.	4.	7.	10.	15.	19.	23.	25.		
Ethyl esters of tall oil fatty acids	30 cal %	83.8	1.1	7.7	19.7	33.2	40.5	50.3	73.0	80.0	0/10	219.4
Ethyl esters of 5,9,12-octadecatrienoic acid	30 "	81.3	−6.3	−4.3	0	2.8	5.0	10.6	16.4	19.3	0/10	157.5
Ethyl esters of linseed oil fatty acids	30 "	84.4	5.4	15.8	30.2	43.9	65.9	77.2	96.6	106.4	0/10	251.7

Table 26. Effect of Pine Seed Oil on Weight Gain of Young Rats (132)

Type and amount of fat in the diet		Mean initial weight, g	Average cumulative weight gain, g — Day of experiment							Deaths	Average food consumption, g
Experiment I			2.	5.	7.	10.	12.	14.	16.		
Pine seed oil	30 cal %	47.0	7.9	21.2	29.3	44.1	54.7	63.7	73.1	0/10	167.2
Soybean extract	30 "	46.8	9.3	25.8	36.4	52.5	63.7	73.4	82.8	0/10	181.9
Soybean oil	30 "	47.2	8.4	23.1	33.0	49.0	60.8	71.7	82.2	0/10	181.1

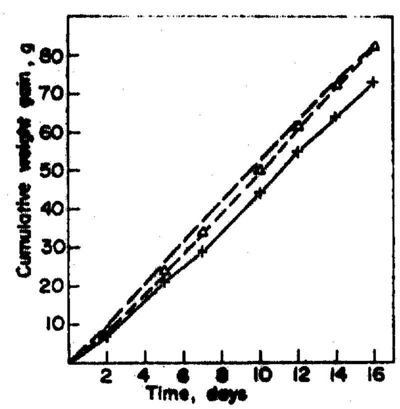

Figure 9. Cumulative Growth Curves for Rats Fed Pine Seed Oil, Soybean Oil and Soybean Extract. (132)

—x—x—x— 30 cal % pine seed oil, **13.2% by weight**
—△—△—△— 30 " soybean oil, **13.2% by weight**
— — — — 30 " soybean extract, **13.2% by weight**

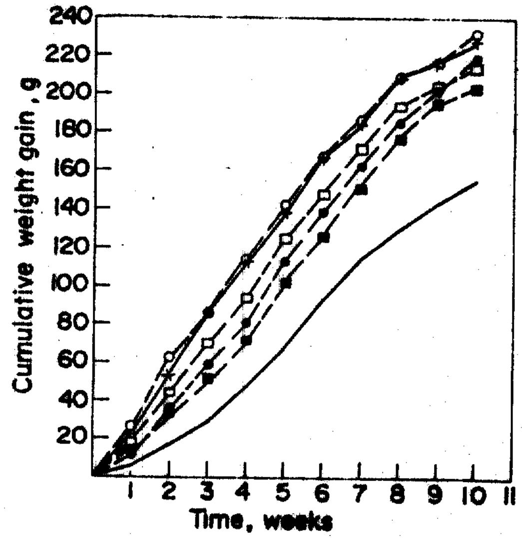

Figure 11. Cumulative Growth Curves for Male Rats Fed Tall Oil Fatty Acid Glyceride Margarine, Butter and Margarine. II Generation (132)

—x—x—x— 30 cal % tall oil fatty acid glyceride margarine , 13.2% by weight
———— 60 » » » » » » , 34.8% by weight
—o—o—o— 30 » butter , 13.2% by weight
—●—●—●— 60 » » , 34.8% by weight
—□—□—□— 30 » margarine , 13.2% by weight
—■—■—■— 60 » » , 34.8% by weight

Table 30. The Effect of Tall Oil Fatty Acid Glyceride Margarine on Weight Gain (132)

Type and amount of fat in the diet		Mean initial weight, g	Average cumulative weight gain, g					Deaths	Average food consumption (7 days), g
			Week of experiment						
			1.	2.	3.	4.	5.		
Males									
Tall oil fatty acid glyceride margarine	30 cal %	38.9	10.4	29.3	52.3	79.2	103.9	0/10	60.7
Tall oil fatty acid glyceride margarine	60 »	37.5	1.1	11.0	15.4	24.2	33.8	0/10	35.6
Butter	30 »	44.1	29.4	70.2	104.9	140.6	176.5	0/10	109.2
Butter	60 »	44.6	23.0	55.5	82.8	121.5	162.9	0/10	81.6
Margarine	30 »	41.4	29.4	72.9	105.2	140.2	171.6	0/10	107.3
Margarine	60 »	49.5	18.4	51.2	75.9	109.4	144.2	0/10	88.8
Tall oil fatty acid glyceride margarine + vit. B	60 »	38.2	6.0	18.0	39.2	54.8	64.4	0/5	40.5
Refined tall oil fatty acid glyceride margarine	60 »	39.4	16.2	26.6	41.8	55.8	64.0	0/5	39.3
Refined tall oil fatty acid glyceride marg. + vit. B	60 »	39.8	19.0	30.8	52.8	72.2	82.4	0/5	46.3
Females									
Tall oil fatty acid glyceride margarine	30 cal %	39.5	12.7	33.2	51.0	68.3	79.4	0/10	62.6
Tall oil fatty acid glyceride margarine	60 »	31.5	3.8	11.0	14.6	21.2	28.0	0/10	28.0
Butter	30 »	39.5	25.8	53.9	74.0	96.9	108.6	0/10	87.0
Butter	60 »	45.6	19.0	46.4	62.2	88.2	108.2	0/10	71.3
Margarine	30 »	39.5	22.5	50.1	73.4	94.6	104.7	0/10	93.0
Margarine	60 »	46.5	18.3	40.2	64.1	85.7	104.6	0/10	82.3
Tall oil fatty acid glyceride margarine + vit. B	60 »	34.0	7.4	17.0	36.4	54.4	64.0	0/5	36.0
Refined tall oil fatty acid glyceride margarine	60 »	35.8	10.2	23.2	39.0	52.2	59.4	0/5	41.8
Refined tall oil fatty acid glyceride marg. + vit. B	60 »	34.0	16.4	28.2	45.2	61.0	70.8	0/5	44.9

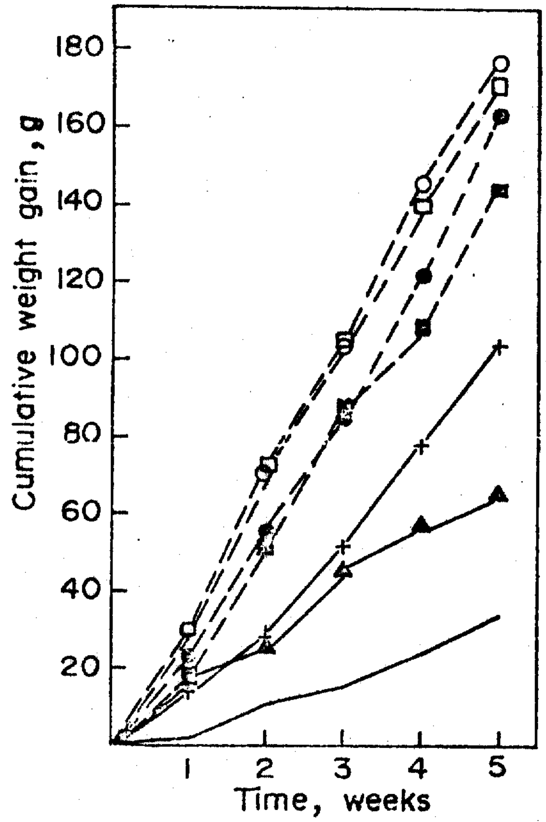

Figure 12. Cumulative Growth Curves for Male Rats Fed Tall Oil Fatty Acid Glyceride Margarine, Butter and Margarine. III Generation(132)

—x—x—x— 30 cal % tall oil fatty acid glyceride margarine, **13.2% by weight**
———— 60 » » » » » » », **34.8% by weight**
—▲—▲—▲— 60 » refined tall oil fatty acid glyceride margarine, **34.8% by weight**
—o—o—o— 30 » butter, **13.2% by weight**
—●—●—●— 60 » », **34.8% by weight**
—□—□—□— 30 » margarine, **13.2% by weight**
—■—■—■— 60 » », **34.8% by weight**

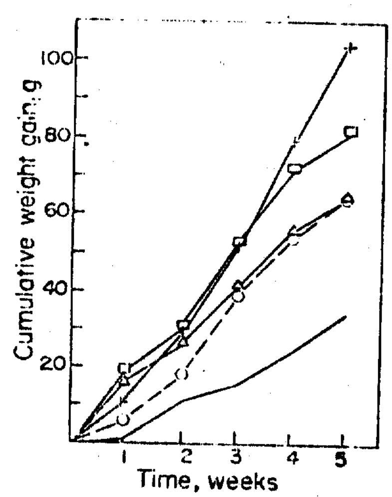

Figure 13. Cumulative Growth Curves for Male Rats Fed Refined Tall Oil Fatty Acid Glyceride Margarine with or without Vitamin B Supplements. III. Generation. (132)

—x—x—x 30 cal % tall oil fatty acid glyceride margarine, **13.2% by weight**
———— 60 » » » » » » », **34.8% by weight**
—o—o—o— 60 » » » » » » » + B vitamins, **34.8% by weight**
—△—△—△ 60 » refined tall oil fatty acid glyceride margarine, **34.8% by weight**
—□—□—□ 60 » » » » » » » » + B vitamins, **34.8% by weight**

In this experiment, the weight gains of all groups, both sexes, on the tall oil margarine were less than those of the controls. Food consumption was recorded for one week only but it seemed to correspond well with growth rates of the various groups. The data obtained indicated that refining the tall oil margarine, coupled with the use of B vitamin supplement, improved the growth-promoting properties.

The main differences, statistically, were as follows:

a. Tall oil margarine compared with butter at lower fat level (males) and butter at higher fat level males & females) was ($P < 0.001$).

b. Tall oil margarine compared with refined tall oil margarine and tall oil margarine + B vitamin supplement was ($P < 0.001$).

E. Rabbits

Weil <u>et al</u>. (158) applied several epoxidized tall oil preparations to the skin of albino rabbits (strain, sex, age, and weight not specified) in graded amounts (undiluted or solutions in water, propylene glycol or acetone). In a 10-grade ordinal series based on the severest reaction that develops from 0.01 ml of the undiluted sample, they then estimated the degree of skin irritation that developed within 24 hours. The results, presented in Table 31, indicate that the tall oil preparations were only slightly irritating to the skin, although the precise effect on one (grade 3) was not completely defined.

Table 31. Effect of Some Epoxidized Tall Oil Preparations on the Skin (158)

Substance	Animals	Sex & No.	Route	Degree of primary[a] skin irritation
Epoxidized 2-ethylhexyl ester of tall oil fatty acids	Albino Rabbits	5	skin	3
Alkyl epoxytallate	Albino Rabbits	5	skin	2
Epoxidized Carbowax 200 ester of tall oil fatty acids	Albino Rabbits	5	skin	2

[a] Degree of primary skin irritation in a 10-grade ordinal series

Grade 1 - No irritation
Grade 2 - "least possible capillary injection from undiluted chemical"
Grade 3 - Not defined completely
Grade 6 - Necrosis from undiluted substance
Grade 10 - Necrosis from 0.01% solution

Weil *et al.* (158) also instilled graded doses of these epoxidized tall oil products in the eyes of rabbits (strain, numbers, sex, etc. not given) and recorded the degree of corneal injury. In each case the degree of corneal necrosis was at most a very small area of necrosis resulting from 0.5 ml of undiluted chemical in the eye.

F. Dogs

Young beagle dogs (number, sex, weight not given) were fed pale tall oil, gum, and wood rosin in the diet at levels of 1.0% and 0.05% for a period of two years (177). Observations during the period included food intake, body weight, gross symptoms, mortality, hematology, urinalysis, liver and kidney function tests, and tumor incidence. At the conclusion of the experiment, all animals were autopsied and examined for gross effects and histopathologic lesions.

At the 1% dietary level, liver enlargement was detected at autopsy in animals on all three rosin products. Microscopic examinations, however, failed to reveal any histologic abnormalities in the liver or other organs attributable to ingestion of the rosins. The weight of all other organs were comparable to those of the controls (kidneys, spleen, heart, brain, gonads, adrenals, and thyroid). No significant differences were noted in food intake and growth rates between test and control animals. Hematology, urinalysis, and liver and kidney function test values were within normal limits throughout the study.

At the 0.05% level, no significant differences were detected between animals on the rosin diets and the controls.

G. Cattle

Antila *et al.* (015) investigated the effect of including tall oil fatty acid ethyl esters in the diet on milk fat composition. This work was carried out in connection with the general problem of the inconsistency of Finnish butter at certain times of the year.

1. In the first experiment, four cows (strain, age, etc. not given) were fed tall oil fatty acid ethyl esters at a level of 4% in the diet over a period of 40 to 45 days[a]. A control group of the same size received the basal diet alone. The yield of milk, milk fat, and protein, as well as various properties of the milk fat, were determined. The results are presented in Figures 14-17 and Tables 32 and 33.

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[a] The experiment extended over a preliminary period of 20-25 days, a transition period of five days, an experimental period of 25-30 days, a transition period of five days, and a final period of five days.

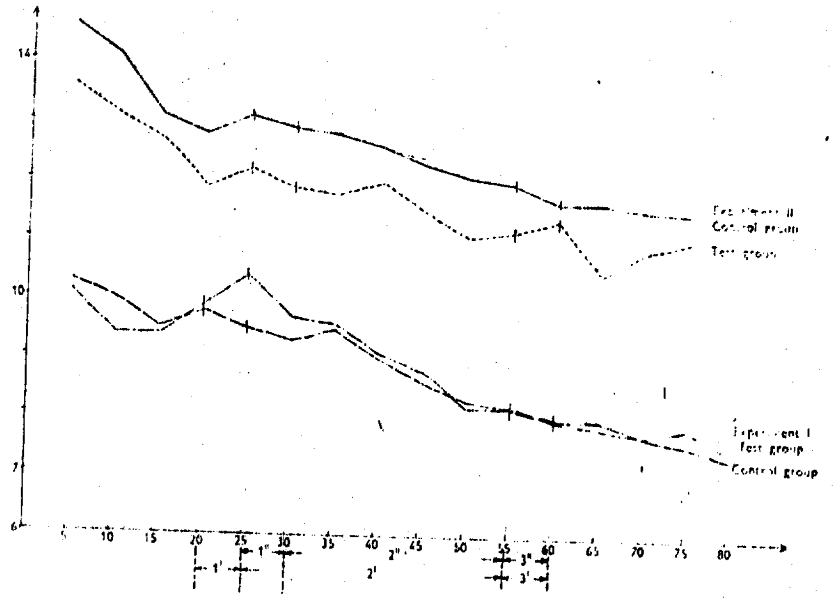

Fig. 14. Variation in Milk Production During the Experiments. 1=Transition period. 2=Test period. 3=Transition period. (015)

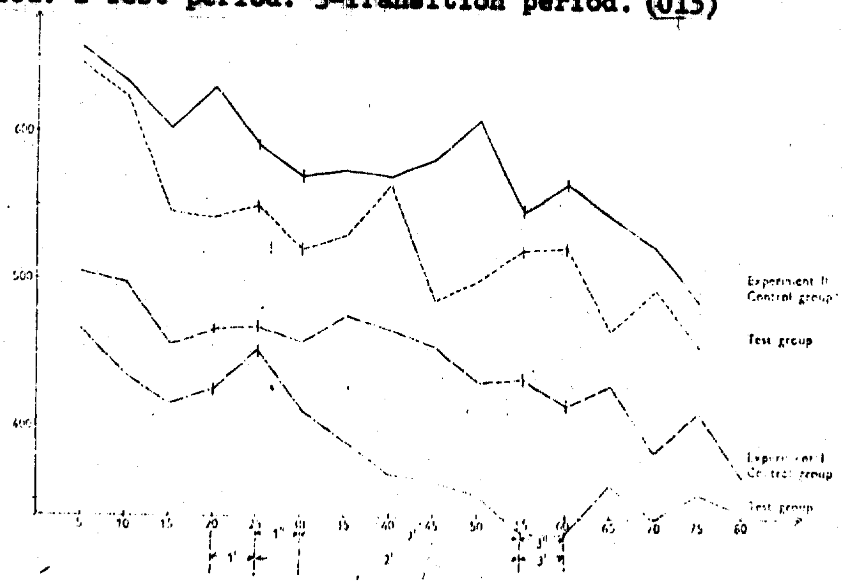

Fig. 15. Variation in Milk Fat Production During the Experiments. 1-Transition Period. 2=Test period. 3=Transition period. (015)

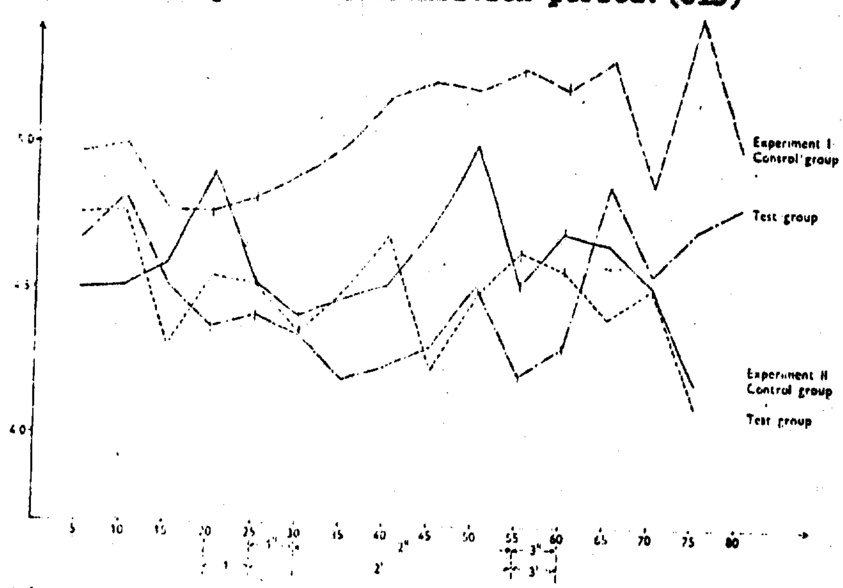

Fig. 16. Variation in Fat Content of Milk During the Experiments. 1=Transition period. 2=Test period. 3=Transition period. (015)

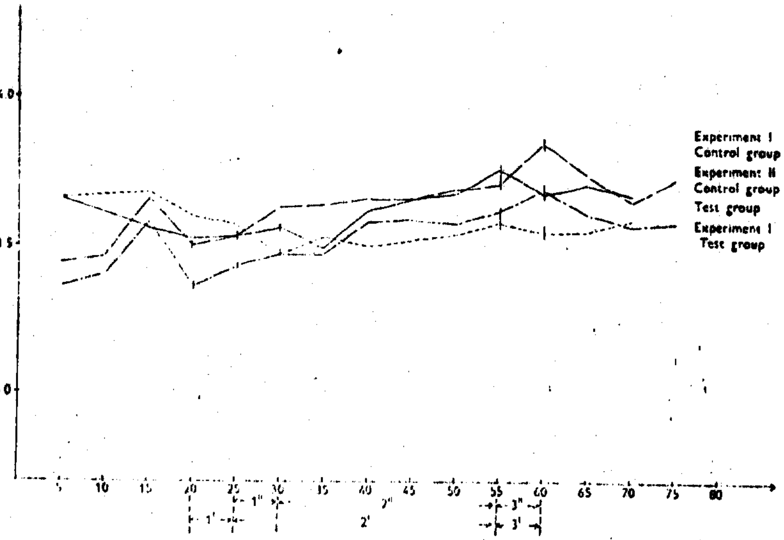

Fig. 17. Variation in Protein Content of Milk During the Experiments. 1=Transition period. 2-Test period. 3=Transition period. (015)

Table 32. Changes in the Iodine Value of Milk Fat Produced by Feeding Ethyl Esters of Tall Oil Fatty Acids to Cows. (015)

	Feeding experiment I		Feeding experiment II	
	Test group	Control group	Test group	Control group
Preliminary period	31.9	30.4	30.2	31.3
Test period	38.1	31.4	33.0	31.8
Follow-up period	33.5	31.3	31.8	33.8

Table 33. The Effect of Ethyl Esters of Tall Oil Fatty Acids in Fodder on the Content of Diethenoids in Milk Fat. (015)

	Feeding experiment I				Feeding experiment II			
	Test group		Control group		Test group		Control group	
	% conjugated	% non-conjugated	% conjugated	% non-conjugated	% conjugated	% non-conjugated	% conjugated	% non-conjugated
Preliminary period	0.63	1.03	0.57	1.04	0.57	1.22	0.58	1.21
Test period	1.02	0.98	0.59	0.98	0.62	1.07	0.55	1.18
Follow-up period	0.65	0.99	0.58	0.88	0.58	1.14	0.64	1.01

Although the milk yield and protein content were not affected significantly (see Figs. 14 and 17), the amount of milk fat was considerably lower in the test than in the control group ($P < 0.01$), as indicated in Figure 15. The investigators called attention, however, to the large daily variation in fat content (see Fog. 16) which may have obscured changes in milk fat production during the experiments.

The iodine value of the milk fat increased significantly ($P < 0.01$) as a result of the tall oil dietary supplement (see Table 32). The investigators point out that this increase not only is statistically significant, but also is of great practical importance since a difference of 1.6 iodine value units is very definitely manifested in the cutting consistency of Finnish butter. In another experiment, Antila discovered that feeding tall oil fatty acid ethyl esters leads to an increase in the oleic acid content and a decrease in palmitic acid content of the fat. He concluded that this effect is responsible for the increased iodine value in the present experiment.

The proportion of conjugated diethenoids of the butter fat also increased significantly ($P < 0.01$) as a result of feeding the tall oil esters (see Table 33.)

2. In the second experiment performed by Antila et al. (015), four cows were fed tall oil fatty acid ethyl esters at a level of 3% in the diet for a period of 30-35 days (first transition period and experimental period). A control group of equal number received the basic diet only. Milk production, fat and protein content, and various properties of the milk fat were determined as before. The results obtained are presented in Figures 14 and 17 and Tables 32 and 33, in comparison with results of the first experiment.

As in the first experiment, milk production and the protein content were not affected (see Figs. 14 and 17). The yield of milk fat was not affected in the second experiment but, as before, its iodine number increased significantly in the group fed the tall oil esters ($P < 0.01$). The proportion of conjugated diethenoids did not increase significantly in the second experiment in contrast to the results of the previous test.

H. Humans

According to the Merck Index, tall oil is a mild local irritant and sensitizer. (139). Systemic toxicity has not been determined (126, 139).

Abietic acid is slightly to moderately toxic and irritating to the skin and mucous membranes (061, 139). It is moderately toxic by ingestion. In mice toxicity by the intravenous route is low but it causes paralysis in frogs (126).

Linoleic acid administered locally can cause mild irritation. Large doses taken orally cause nausea and vomiting (139). In rats, large doses cause weight loss, progressive anemia, leucopenia, and pediculosis (126).

Oleic acid has low oral toxicity but is mildly irritating to the skin and mucous membranes. (126,139).

Rosins (resin acids) have a slight local toxic effect (126). Systemic toxic effects are estimated to be slight (139).

β-sitosterol from tall oil is used as an anti-cholesterolemic agent in human medicine. Divided doses totaling as much as 30 grams a day are prescribed. Untoward effects apparently vary with the patient. Anorexia, gastrointestinal cramps, and diarrhea have been observed in some patients receiving very large doses, whereas others on the medication for as long as 18 months have experienced no serious side effects (112,125,140).

III. Long-term Studies

A. Mice

Weil et al. (158) recorded mortality at various intervals during the carcinogenicity study reported later in this section (see Biological Data IV, A, 1). The results obtained are summarized in Table 34.

Table 34. Long-term Effect of Some Epoxidized Tall Oil Products on Mice (158)

Substance	Animals	Sex & No.	Route	Dosage	Number of mice alive at month 12	17	24
Epoxidized 2-ethylhexyl ester of tall oil fatty acids	C3H Mice	30-40	Skin	Undil., one brushful, 3/wk for life	30	23	6
3,4-epoxy-6-methyl cyclohexylmethyl ester of tall oil fatty acids	C3H Mice	30-40	Skin	Undil., one brushful 3/wk for life	36	30	3

B. Rats

1. Seppanen (132) studied the effect of tall oil fatty acid glyceride margarine on longevity with rats left over from the reproduction experiment (see Biologicat Data, II,D, 12 and IV, B,1).

Male and female rats, seven to nine animals of the same sex per group were fed tall oil fatty acid margarine at a level of 13.2% in diet type D for the remainder of their life span. Control groups of the same number and sex received either butter or regular margarine. The animals were weighed onee a month and fatalities were recorded; survival and growth curves were plotted from these data. The results are presented in Figures 18-20.

Female rats fed tall oil fatty acid glyceride margarine had much longer life spans than those of the butter or commercial margarine control animals. Male rats on the tall oil product had life spans almost as long as those on commercial margarine and considerably longer than those on butter.

2. In a long-term study at Hercules (177), young albino rats (strain, sex, weight, and numbers not specified) were fed pale tall oil, gum, and wood rosin at levels of 1, 0.2, and 0.05% in the diet for a period of two years. The animals were observed throughout for toxic symptoms, mortality, food consumption, and growth rate. In addition, urine and hematologic analyses, and liver and kidney function tests were performed. All animals were autopsied at the end of the feeding period and examined for gross and microscopic abnormalities, including tumor incidence.

At the 1% dietary level, enlarged livers were noted in the animals on all three rosin diets. No histopathologic abnormalities were detected, however, either in the enlarged livers or in any other of the organs studied (heart, brain, kidneys, spleen, stomach, small intestine, colon, bladder, pancreas, lungs, muscle, prostate, uterus, gonads, adrenals, lymph nodes, thyroid, and parathyroid glands). All other organ weights of test animals were approximately the same as the controls. Hematology, urinalysis, and liver and kidney function test values were within normal limits throughout the study. However, with all three rosin products, food consumption was approximately 10% less than that of the controls, resulting in a slight depression of the growth rate.

At the 0.2% and 0.05% levels, no significant differences were detected between test and control animals.

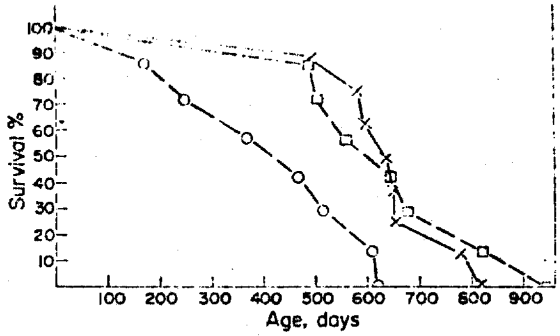

Figure 18. Survival Curves of Male Rats Fed Tall Oil Fatty Acid Glyceride Margarine, Butter and Margarine. (132)

—x—x—x— 30 cal % tall oil fatty acid glyceride margarine
—o—o—o— 30 » butter
—□—□—□— 30 » margarine

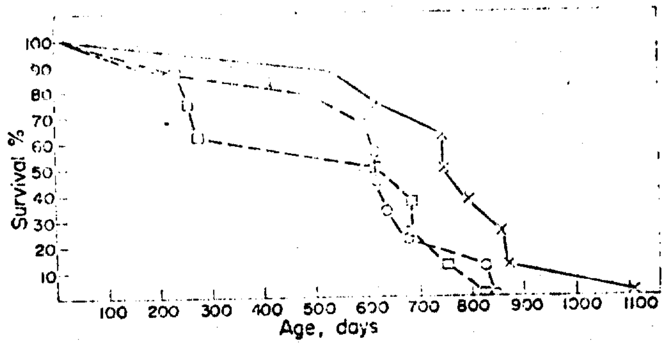

Figure 19. Survival Curves of Female Rats Fed Tall Oil Fatty Acid Glyceride Margarine. Butter and Margarine (132)

—x—x—x— 30 cal % tall oil fatty acid glyceride margarine
—o—o—o— 30 » butter
—□—□—□— 30 » margarine

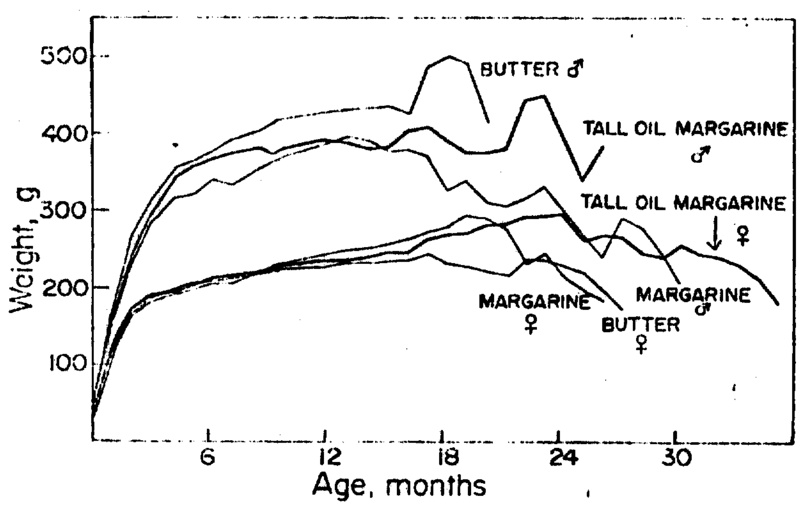

Figure 20. Growth of Rats Fed Tall Oil Fatty Acid Glyceride Margarine, Butter and Margarine at a level of 30% of Dietary Calories in Longevity Experiment. (132)

IV. Special Studies

A. Mice

1. Weil et al. (158) assayed two epoxidized tall oil preparations (epoxidized 2-ethylhexyl ester and 3,4-epoxy-6-methylcyclohexylmethyl ester, of tall oil fatty acids) for carcinogenicity potential. This was part of an investigation involving a large number of representative epoxides which were being studied for commercial utility.

C3H mice, 90 days of age, (sex and weight not given), in groups of 30 to 40, were painted with the compounds (undiluted) on the clipped skin of the back (midline area[a]). One brushful was applied three times each week (M,W,F) for 27 to 28 months; the animals were observed for papillomas and carcinomas at each painting period.

Neither of the compounds gave any evidence of igenicity under the conditions of the study.

2. Albino mice (age, sex, number not given) injected intraperitoneally with 0.02 ml of oxidized oleic acid manifested chromosome aberrations in bone marrow cells (111).

B. Rats

Seppanen (132) studied the influence of tall oil fatty acid glyceride margarine on reproduction in the rat in a series of three experiments with animals remaining from one of the short-term growth experiments (see Section II, D, 12).

1. In the first reproduction experiment, ten female rats and five males from each group of the above-mentioned experiment were fed tall oil fatty acid glyceride margarine at levels of 13.2% and 34.8%. Control groups of the same number were placed on butter or commercial margarine. The rats were 100-200 days old.

All males were transferred to the next cage of the same experimental group after one week of breeding. After 14-16 days, the males were removed permanently. The females were placed in separate cages before the 21st day after the original mating. The offspring were weaned 21 to 28 days after birth depending on the weight attained. The number of litters, offspring, fatalities, and victims of cannibalism were recorded. Results are given in Table 35.

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[a] Hair was removed with electric clippers

Table 37. **Reproduction Experiment III. Number of Litters and Young in Different Groups and Mortality Percentages** (132)

Type and amount of fat in the diet	Number of females	Litters	Total number of young	Mean	Range	Number of dead young	Mortality %	Eaten
Tall oil fatty acid								
glyceride margar. 30 cal %	5	5	50	10.0	8–12	10	20.0	6
» » » 30 » +B[1]	5	5	64	12.8	9–16	40	62.5	28
» » » 30 » +E[2]	5	5	47	9.4	3–13	25	53.1	21
» » » 60 »	5	4	33	8.3	6–14	33	100	11
» » » 60 » +B	5	5	39	7.8	7– 9	11	28.2	6
» » » 60 » +E	5	0	—	—	—	—	—	—
Butter 30 »	5	4	36	9.0	3–14	8	22.2	7
» 30 » +B	5	5	62	12.4	9–14	20	32.3	10
» 30 » +E	5[3]	4	45	11.3	8–12	10	22.2	4
» 60 »	5	3	21	7.0	2–10	8	38.1	7
» 60 » +B	5	5	62	12.4	9–17	10	16.1	4
» 60 » +E	5	3	21	7.0	2–12	11	52.4	3
Margarine 30 »	5	4	38	9.5	7–12	4	10.5	3
» 30 » +B	5[3]	5	50	10.0	5–12	25	50.0	17
» 30 » +E	5[4]	5	32	6.4	3–12	10	31.2	4
» 60 » +B	5	5	41	8.2	4–11	4	9.7	3
» 60 » +E	5	5	47	9.4	3–12	24	51.1	16

B = vitamin B-supplement.
E = vitamin E-supplement.
One female died.
Two females died.

C. Rabbits

1. Several workers have produced tumors in rabbits within 4-12 weeks by injecting oleic acid subcutaneously (150).

2. Altschul (003) fed rabbits (age, sex, numbers not given) stigmasterol as a level of 0.3 grams daily for 73-116 days in a carcinogenicity study. No tumors developed (003).

D. Barley

Bhatnagar _et al_. (021) treated huskless barley seeds with oleic acid for 4-12 hours and noted that there was a reduction in pollen fertillity of the M2 plants, a marked increase in frequencies of chromosome bridges and fragments, and a low frequency of chlorophyll mutations.

BIOCHEMICAL ASPECTS

I. Breakdown

During storage of wood for pulping, the fat and resin constitutents undergo oxidation or other modifications resulting in a decreased yield of tall oil. In one study, during a twelve-week period, the loss amounted to 11% in the case of pine roundwood and 64% for wood chips. The decreased yield from roundwood was entirely from a loss of fatty acids. Fatty acid loss accounted for most of the decreased yield from chips also, but in addition, there was a small loss in resin acids and a very small loss of unsaponifiable matter. The quality of tall oil also suffered from changes during the storage period as the acid number dropped below the minimum specification of 160 (034,132).

According to Seppanen (132), the elevated temperatures of the sulfate pulping process may bring about changes in the highly unsaturated fatty acids of tall oil and result in the formation of toxic polymers and cyclic monomers.

Abietic acid, the main resin acid in tall oil, is unstable (at least under certain circumstances) apparently due to its double bond structure. Disproportionation, or decreasing the number of double bonds, is done commercially and makes for a more stable product. Disproportionated rosin soaps are used, for example, as emulsifiers in the manufacture of styrenebutadiene synthetic rubber (007).

Tall oil and its products darken on standing and should be stored in containers of aluminum or 18-8 stainless steel. Tank cars and drums should be lined with one or the other of these substances. A blanket of inert gas should be layered over the product to reduce the oxidative changes.

Antila et al. (015) investigated the stability of tall oil fatty acid ethyl esters as supplements in cattle feed held in the dark at room temperature over a period of eight weeks. Measurement of oxidative changes in the ester product and in mixtures with various feed components were followed by determination of peroxide values. The results indicate that the ester product was oxidized quite rapidly when mixed with feed, probably a result of the greater exposure to air as compared with the unmixed control. Addition of grass meal, which contains anti-oxidants, stabilized the preparation quite well.

Seppanen (132) concluded that the tall oil fatty acid glyceride margarine used in his study became oxidized to a certain extent and that oxidation products caused partial decomposition of certain B vitamins in the experimental diets.

Since the subject of this monograph is tall oil, and since fats and their components have been covered in at least one other monograph, no attempt is made here to summarize the voluminous literature on that subject.

II. Absorption-Distribution

Seppanen (132) investigated the absorbability in rats of tall oil fatty acid distillate (I), ethyl esters of distilled tall oil fatty acids (II), and tall oil fatty acid glyceride margarine (III).

Sprague-Dawley animals were used throughout (age, sex, weight, etc. are given in Tables 38 and 39). I and II were fed at a level of 13.1% in diet type B. III was fed at levels of 13.1% and 34.6% in diet types B and C, respectively. Soybean oil fatty acids, soybean oil and butter were employed as control fats. The experimental period varied from five to 20 days. Other details of the experiment and results obtained are presented in Tables 38 and 39.

As indicated in these tables, the absorbabilities of tall oil fatty acids (96-97%) and tall oil fatty acid glyceride margarine (95-96%) were relatively high. The ethyl esters of distilled tall oil fatty acids had the lowest absorption percentage (93%), which was 5% less than that of the corresponding soybean oil controls. The tall oil fatty acids distillate was absorbed almost as well as the soybean oil preparation (1-2% less). The tall oil fatty acid glyceride margarine was absorbed better than butter in most tests, the variation being from 2% to 8%.

III. Metabolism and Excretion

No direct information was found in the literature on the metabolism and excretion of tall oil or its fatty acid components. Apparently the unsaturated fatty acids are metabolized by the same route as those from other vegetable oils.

IV. Effects on Enzymes and Other Biochemical Parameters

Seppanen (132) determined the effect feeding tall oil fatty acids at a level of 13% had on the lipid composition of various tissues in the rat. Animals from the absorption experiment were used (see this section, II).

The animals were usually fasted overnight before anesthetization (ether ad. narcosin) and removal of tissues to be analyzed. Blood plasma was stored in the refrigerator until analyzed a few days later. The liver was blotted dry, weighed, cooled immediately and stored in an air-tight glass jar at dry ice

Table 38. **Absorbability of Fat in Young Rats Given Tall Oil Fatty Acid Glyceride Margarine and Butter[a] (132)**

Type and amount of fat in the diet			Sex of rats[b]	Method of determination		Number of samples	Mean absorbability percentage
Tall oil fatty acid glyceride margarine	30	cal %	male	chromium oxide[c]		10	95.7±0.82[d]
Tall oil fatty acid glyceride margarine	30	»	female	»	»	10	93.9±0.67
Tall oil fatty acid glyceride margarine	60	»	male	»	»	10	95.2±0.75
Tall oil fatty acid glyceride margarine	60	»	female	»	»	10	96.4±0.31
Butter	30	»	male	»	»	10	94.3±0.62
»	30	»	female	»	»	10	95.5±0.32
»	60	»	male	»	»	10	92.9±1.15
»	60	»	female	»	»	10	88.0±1.60

a Uncorrected for endogenous fat excretion
b Standard error of mean.
c Chromium determined by colorimetry
d Each group consisted of ten animals weighing 41-44g (initial weight)

Table 39. **Absorbability of Fat in Male Rats Given Tall Oil Fatty Acid Distillate, Ethyl Esters of Distilled Tall Oil Fatty Acids, Soybean Oil and Soybean Fatty Acids.[a] (132)**

Type and amount of fat in the diet			Age of rats	Method of determination	Number of samples	Mean absorbability percentage
Tall oil fatty acid distillate	30	cal %	young[e]	chromium oxide[c]	5	96.7±0.32[b]
» » » » »	30	»	full-grown[f]	» »	5	96.9±0.38
» » » » »	30	»	full-grown[f]	food consumption	25	96.4±0.38
» » » » »	30	»	young[g]	chromium oxide[d]	59	95.7±0.05
Ethyl esters of distilled tall oil fatty acids	30	»	young[e]	chromium oxide[c]	5	93.1±0.96
Soybean oil fatty acids	30	»	full-grown[f]	chromium oxide[c]	5	97.4±0.08
» » » »	30	»	full-grown[f]	food consumption	25	97.6±0.49
» » » »	30	»	young[g]	chromium oxide[d]	60	96.6±0.05
Soybean oil	30	»	young[e]	chromium oxide[c]	5	98.1±0.32

a Uncorrected for endogenous fat excretion
b Standard error of mean
c Chromium determined by colorimetry
d Chromium determined by atomic absorption spectrophotometry
e Five 40g rats (initial wt.)
f Five 250 g rats (initial wt.)
g Ten 95.g rats (intital wt.)

Table 41. Fatty Acid Compositions of Adipose Tissue Triglyceride Fractions From Rats Fed Tall Oil Fatty Acids and Soybean Oil Fatty Acids at a Level of 30% of Dietary Calories (13.2% by weight) (132)

	$<C_{14}$	C_{14}	C_{15}	C_{16}	$C_{16:1}$	C_{17}	C_{18}	$C_{18:1}$	$C_{18:2}$	$C_{18:3}$[a]	$C_{18:3}$	C_{20}	$C_{20:4}$	Other acids
Tall oil fatty acids (6)[b]	1.1	1.4	0.3	18.9	2.1	0.3	2.4	47.4	20.7	1.0	0.6	1.5	0.0	2.2
S.E.[c]	±0.1	±0.1	±0.05	±1.6	±0.6	±0.07	±0.2	±2.0	±4.3	±0.3	±0.08	±0.5	±0.02	±0.05
Soybean oil fatty acids (4)	1.9	1.5	0.6	23.1	11.1	0.1	5.2	44.6	5.8	--	1.3	2.0	0.4	2.5
S.E.	±0.3	±0.2	±0.06	±4.4	±0.8	±0.06	±0.7	±4.1	±1.7	--	±0.3	±1.0	±0.3	±0.4

a. cis-5,9,12-Octadecatrianoic acid
b Number of rats
c Standard error of mean

Table 42. Fatty Acid Compositions of Liver Lipid Fractions from Rats Fed Tall Oil Fatty Acids and Soybean Oil Fatty Acids at a Level of 30% of Dietary Calories (132)

	$<C_{14}$	C_{14}	C_{15}	C_{16}	$C_{16:1}$	C_{17}	C_{18}	$C_{18:1}$	$C_{18:2}$	$C_{18:3}$[a]	$C_{18:3}$	C_{20}	$C_{20:4}$	Other acids
Triglyceride fraction														
Tall oil fatty acids (8)[b]	2.1	0.8	0.6	23.4	1.4	0.5	13.8	10.3	27.4	2.1	0.3	1.0	3.8	3.5
S.E.[c]	±0.4	±0.08	±0.1	±0.8	±0.3	±0.05	±1.8	±1.6	±0.3	±0.07	±0.09	±0.1	±0.7	±0.6
Soybean oil fatty acids (8)	1.5	1.2	2.4	30.6	1.2	0.2	3.3	25.9	29.2	—	0.8	1.5	0.2	1.9
S.E.	±0.4	±0.1	±0.7	±2.4	±0.1	±0.02	±0.6	±1.0	±3.1	—	±0.2	±0.2	±0.09	±0.2
Phospholipid fraction														
Tall oil fatty acids (9)	2.3	0.2	0.5	26.5	0.3	1.2	40.8	9.7	8.2	0.4	0.0	0.1	8.0	1.8
S.E.	±0.5	±0.02	±0.06	±1.2	±0.04	±0.1	±1.1	±0.6	±0.8	±0.05	—	±0.03	±2.2	±0.2
Soybean oil fatty acids (6)	2.9	0.3	0.9	25.4	0.3	0.7	42.6	6.5	10.9	—	0.2	0.8	7.6	0.9
S.E.	±1.0	±0.04	±0.4	±1.7	±0.04	±0.1	±3.3	±0.2	±1.7	—	±0.07	±0.1	±4.5	±0.1

a cis-5,9,12-Octadecatrienoic acid
b Number of rats
c Standard Error of Mean

The major fatty acids of the liver triglycerides in the soybean oil group were palmitic, oleic, and linoleic acids, each amounting to more than one-fourth of the total acids. In the tall oil groups, the fatty acid contents were a few percent lower. However, the percentage of stearic acid in the soybean oil control group was only 3.3% as compared with 13.8% in the tall oil group. The fatty acid composition of the liver phospholipids was very similar in both groups. Over 2% cis-5,9,12-octadecatrienoic acid was found in the liver triglyceride fraction of animals in the tall oil group.

Seppanen (132) fed adult male rats glyceryl esters of tall oil fatty acids at a level of 13.1-13.2% in the diet over a period of three weeks to determine the effect on the composition of the fecal lipids. Diet controls were not included in this experiment. Random fecal samples were collected from ten rats during the feeding period and stored under nitrogen at -20°C until analyzed. The results are given in Table 43.

Table 43. **Fatty Acid Composition of Fecal Lipids from Rats Fed Glyceryl Esters of Tall Oil Fatty Acids at a Level of 30% of Dietary Calories (132)**

	$<C_{14}$	C_{14}	C_{15}	C_{16}	$C_{16:1}$	C_{17}	C_{18}	$C_{18:1}$	$C_{18:2}$	$C_{18:3}$[a]	$C_{18:3}$	C_{21}	$C_{20:4}$	Other acids
Mean percentage S.E.[b]	1.1 ±0.2	0.6 ±0.1	0.8 ±0.08	4.9 ±0.3	1.3 ±0.1	0.7 ±0.1	2.8 ±0.1	25.3 ±1.0	10.7 ±0.8	16.2 ±0.5	8.3 ±0.3	2.6 ±0.4	1.0 ±0.4	14.7 ±0.8

a cis-5,9,12-Octadecatrieonic acid

b Standard error of mean

The major discovery in this experiment was the relatively high content of cis-5,9,12-octadecatrienoic acid (16.2%) found in the fecal lipids. This was considerably higher than the amount found in any of the tissue lipid fractions studied. The predominant fatty acids in the feces were oleic and linoleic acids, each being present in a concentration of 20% or more.

The effects of tall oil products on appetite and growth of rats were given earlier in the discussions of Seppanen's experiments; the changes in milk fat, cheese, and egg yolk resulting from feeding these substances to chickens and cattle were presented in the summaries of Antilla's studies.

V. Drug Interaction

No information on drug interactions was found in the literature, except for Seppanen's conjecture (132) that oxidized tall oil constituents in the tall oil fatty acid margarine used in one of the feeding experiments with rats destroyed some of the vitamins in the basal diet. This fact has been demonstrated in the case of other natural fats and oils (See IV. Special Studies).

VI. Consumer Exposure Information

Whole tall oil is not a direct food additive; consequently, consumer exposure is limited mainly to its presence in certain food packaging and processing articles where it is used as a manufacturing additive.

Certain textiles and textile finishes used in dry food packaging are important articles from which tall oil might possibly migrate to foods - 21 CFR 121.101 (011).

Tall oil may be used in concentrations up to 5% as an emulsifier in the manufacture of certain kinds of rubber items, intended for repeated use, that may be employed in food processing or packaging - 21 CFR 121.2520 (011).

Finally, tall oil or tall oil precursors are apparently discharged into natural bodies of water in certain parts of the world. The mining industry uses large amounts of tall oil and tall oil products as flotation agents in the separation of ore components (119). Tall oil fulfate soap in kraft pulp mill wastes has been found to be detrimental to fish and shellfish in several areas (030,152).

In contrast to the situation with whole tall oil, consumer exposure to tall oil constituents and compounds is quite wide on account of the many commercial applications of these substances (See CHEMICAL INFORMATION, VIII).

The possibility of using tall oil fatty acid glycerides as edible oils has been under consideration for some time -- especially since tall oil is the world's cheapest source of edible fatty acids (007). According to Palonini (113), mixtures of the glycerides of these acids have actually been sold as Grade B olive oil, and have been used to adulterate other food oils.

Two Canadian patents have been granted (132) for edible products prepared from refined tall oil fatty acid glycerides. One of these products, a salad oil, is said to have a flavor and certain other characteristics superior to products made from corn oil and soybean oil. The other products, a shortening and a margarine, are prepared by blending or reacting hydrogenated tall oil fatty acid glyceride margarine with animal fats such as tallow or lard and certain vegetable oils like palm oil, soybean oil, or cottonseed oil. The process results in products with the desired consistency, melting range, and cold test properties.

Oleic acid derived from refined tall oil fatty acids is permitted in foods in the United States if it is free from the chick-edema factor and is in conformance with certain other specifications - 21 CFR 121.1237 (011).

Glyceryl-lacto esters of oleic acid derived from refined tall oil fatty acids which meet certain specifications are permitted in other food grade additives - 21 CFR 121.1004, 1048 (011).

New synthetic chewing gum bases have been developed in which glyceryl esters of refined tall oil rosin are used as plasticizers (softeners) (009).

Some emulsifiers used in the food industry contain lactylic esters of oleic acid derived from refined tall oil fatty acids - 21 CFR 121.1048 (011).

Defoaming agents, which are non-toxic and highly effective, have been developed from tall oil-ethylene oxide condensation products. Some of these are used in yeast production and in the antibiotic industry (104).

Binders and lubricants used in certain areas of the food industry contain glyceryl-lacto esters or salts of oleic acid derived from refined tall oil - 21 CFR 121.1004,1048 (011).

Salts of tall oil oleic acid are employed as anti-caking agents in the manufacture of certain foods - 21 CFR 121.1071 (011).

Glyceryl abietate is used as an emulsifier for citrus oil in still and carbonated alcoholic beverages and fruit drinks in concentrations up to 0.006% (102).

Abietic acid is utilized as a carrier in enriched rice; concentrations up to 0.0026% are permitted (049).

Surface coatings for fresh fruits (up to 0.02%) are prepared from pimaric and abietic acid copolymers (049).

Tall oil rosin, refined to certain specifications, is permitted for use as sizing for paper as well as other articles or components of articles used in producing, manufacturing, packing, processing, preparing, treating, packaging, transporting, or holding food - 21 CFR 121.2592 (011).

Abietic, oleic, linoleic acids may be employed in defoamers for beet sugar products in concentrations up to 0.0001% (049).

Coatings of various types in contact with foods may contain tall oil rosin and demerized fatty acids derived from tall oil which meets certain specifications - 21 CFR 121.2557, 2592 (011).

Tall oil fatty acids and their N-butyl and isooctyl esters are utilized as activators in the manufacture of rubber which may be employed in food processing 21 CFR 121.2562 (011).

A wide variety of tall oil products - rosin, rosin salts, pitch, abietic acid, fatty acids and their methyl, polyoxyethylene, polyethylene glycol (600) esters - are used in adhesives for paper and paperboard food containers - 21 CFR 121.2520 (011).

Tall oil fatty acids and triglycerides, as well as alcohols and dimers derived from tall oil, are used in defoaming agents employed in the manufacture of paper and paperboard containers that find applications in packaging, transporting or holding food - 21 CFR 121.2519 (011).

Tall oil mixed soap (Ca, K, Na), and the glyceryl esters of maleic anhydridemodified tall oil rosin and disproportionated tall oil rosin, are employed also in the manufacture of articles or components of articles that may be used in food processing, packaging, or holding - 21 CFR 121.2562 ( ).

Tall oil fatty acids are being employed in increasing amounts in soap manufacture in place of the more expensive linseed and soybean oils and animal fats (172).

Patents have been granted for the use of methyl esters of tall oil fatty acids, (up to 0.5%) with urea and other substances, in animal feeds.

Tall oil sterols are used in several areas of human medicine. β-sitosterol from tall oil is employed as an anti-cholesterolemic agent. (112,125,140).

Tall oil sterols are used in Sweden also for the manufacture of sex hormones and other steroid products (006).

No information was found in the literature on the average daily intake of tall oil or any of its constituents excepting β-sitosterol. According to Salen (121), the average American dietary intake of this plant sterol yields a plasma concentration of 0.30 to 1.02 mg/100 ml. Absorption is 5% or less of the daily intake as compared to 45-54% for cholesterol. About 20% of the absorbed -sitosterol is converted to cholic acid and chenodeoxycholic acid; the remainder is excreted in the bile as the free sterol. It is excreted more rapidly than cholesterol. There is no endogenous synthesis of β-sitosterol.

The world production of crude tall oil is estimated to be about 1500 million pounds per year of which the United States alone produces approximately 1000 million pounds (132). In 1972, $34,025 worth of tall oil was imported from Canada, and $1,176 worth from France (149).

The price of tall oil in tank car lots is 3.8¢ per pound for the crude grade and 8.5¢ for the refined oil. This low price accounts to a large measure for the increasing industrial use of tall oil and its products (148).

APPENDIX A

I. Nomenclature

The name tall oil is the American equivalent of the sound of the German Talloel which in turn comes from the sound of the original Swedish name Tallolja (Tall - pine; olja - oil)

The following synonyms for tall oil should be considered obsolete but some are still used: Finn oil, liquid resin, liquid rosin, resin oil, sulfate pitch, sulfate resin, sulfate rosin, Swedish pine oil, Swedish rosin, Swedish rosin oil, Sylvic oil, Talloel, and Tallol (161).

II. Some Manufacturers of Tall Oils

Crude Tall Oil

Company	Trade Name
Arizona Chemical Co.	Acintol C
Camp Mfg. Co.	Campol No. 8485
Champion Paper & Fiber Co.	Trostol
Continental Can Co., Hummel-Ross Div.	Superior
National Southern Products Corp.	Opoil
North Carolina Pulp Co.	Plymouth
Southern Advance Bag & Paper Co.	Advanol
Union Bag & Paper Corp.	
West Virginia Pulp & Paper Co.	Ligro; Covoil

Distilled and Refined Tall Oils

Arizona Chemical Co.	Distilled	Acintol D; Acintol DLR
Camp Mfg. Co.	Refined	Campol Nos. 36S;36V;792S; 792V
Gaylord Container Co.	Refined	Refined Bogol
Hercules, Inc.	Distilled	Pamaks-W10,15,25A,45A,25, 25A;LE,DLE
National Southern Products Corp.	Refined	Facoil CS,CP,CB,CU,CN
Newport Industries, Inc.	Distilled	Acolin; Acosix; Aconon
Union Bag & Paper Co.	Refined	Unitol S,R,V
West Virginia Pulp & Paper Co.	Single Vacuum Dist.	Rosoil "AH"
	Double distilled	Indusoil
	Triple distilled	Indusoil

III. Some American Commercial Tall Oils

The composition and properties of some American commercial tall oils and tall oil products are given in the following tables.

Table 44. Typical Propoerties of PAMAK Tall Oil Products[a]

	Fatty acids %	Rosin acids %	Unsaponi-fiables %	Color (Gardner)	Acid number	Iodine number	Titer, °C.
FATTY ACIDS							
Pamak 985	98.7	0.6	0.7	<1	197	129	3
Pamak 1	98.4	0.7	0.9	1-2	197	130	5
Pamak 4A	95.2	3.0	1.8	3+	193	132	4
Pamak W4A	96.0	1.8	2.2	4	194	132	3
Pamak 4	92.5	3.5	4.0	6	190	130	5
Pamak W4	92.0	4.0	4.0	6+	190	130	0
Pamak 6	90.5	7.0	2.5	6	190	130	5
Pamak WD	87.1	1.7	10.8	6+	176	128	<1
DISTILLED TALL OILS							
Pamak W10	86.0	10	4.0	7	187	131	-
Pamak W15	81.5	15	3.5	8	185	131	-
Pamak 25	70.7	27	2.3	8	184	137	-
Pamak 25A	70.2	28	1.8	6+	187	140	-
Pamak W25A	70.0	26.2	2.5	7	188	137	-
Pamak W45A	52.5	43	4.5	7	175	135	-
TALL OIL LIGHT ENDS							
Pamak LE	74-76	0.8-3.3	22-27	12-16	151-159	-	-
Pamak DLE	84.8	0.5	14.7	11	175	-	-
Pamak WLE	55.0	1.5	40	13	120	-	-

[a]Courtesy of Hercules, Inc., Wilmington, Delaware

Table 45. Tall Oils (161)

Commercial Name of Product	Produced by	Acid Number Min.	Acid Number Max.	Iodine Number Min.	Iodine Number Max.	Saponification Number Min.	Saponification Number Max.	Unsaponifiable Min.	Unsaponifiable Max.	Specific Gravity 15.5° C./15.5° C. Min.	Specific Gravity 15.5° C./15.5° C. Max.	Refractive Index at 25.0° C.	Color Gardner 20°-30° C.	Absolute Viscosity at 25° C. Gardner-Holdt	Absolute Viscosity at 25° C. Poises	Composition Fatty Acids %	Composition Rosin Acids %
★ Distilled	Albemarle	170		140				6					8-10	E-F		59	35
Crude	Arizona	160	175	160	180	165	180	6.0	8.0	0.97	0.99		Brown			47	45
Crude	Champion	160	170			165	175	5.0	8.0							45-52	38-45
1103 Refined	Dorward		165										11	X-Y			
Bogol	Gaylord	165	180			165	180	4.5	5.5							56-60	34-40
Aliphat 45-B	General		175													30	70
• Crude	Hummel-Ross	163	170			165	185	7.5	10.0							45-53	36-45
Opoil	N.S.P.		171.9		159.1		173.9	8.0					Brown			49.5	42.2
Deoil	N.S.P.		171.9		159.1		173.9	8.0					Brown			49.5	42.2
Facoil CB	N.S.P.		169.0		155		177.5	7.5			0.973		10-12			50.0	41.5

Table 46. Fatty Acid Composition of ACINTOL Tall Oil Fatty Acids and Selected ACINTOL Distillation Products by Gas Liquid Chromatography[a]

								ACINTOL DISTILLATION PRODUCTS		
	D6LR	FA-1	FA-1 Special	FA-2	FA-3	EPG	B29LR	D30E	DLR Special	Heads 2122
Lower Boiling Acids, %	0.1	—	—	—	—	—	—	—	—	8.5
Palmitic Acid, %	5.3	2.3	1.9	0.1	0.1	—	1.9	—	1.6	35.9
Palmitoleic Acid, %	2.1	0.6	1.2	0.2	0.1	—	1.0	—	0.8	8.5
Unknown Acid, %	0.3	0.3	0.1	0.1	0.1	—	0.1	—	0.4	1.4
Stearic Acid, %	2.1	2.3	2.2	2.5	2.6	2.5	1.6	1.2	2.0	1.0
Oleic Acid	38.3	41.0	43.0	49.5	48.0	49.6	38.6	10.0	31.5	23.1
Unknown Acid, %	1.6	1.2	1.7	1.6	2.0	1.3	1.5	0.7	2.0	0.9
Linoleic Acid (cis-9, cis-12), %	29.4	32.8	32.2	35.7	38.1	36.5	28.1	9.2	24.3	17.8
Unknown Acid, %	2.3	2.5	2.5	3.1	2.7	2.8	2.8	0.8	2.4	1.9
Unknown Acid, %	0.9	0.6	0.9	0.4	0.2	0.2	0.7	1.0	1.8	0.1
Linoleic Acid (cis-9, trans-11), %	4.3	3.8	4.4	2.6	2.4	2.5	4.2	13.3	7.1	0.4
Eicosanoic Acid, %	1.7	1.9	1.8	1.4	1.2	1.3	1.8	4.3	1.8	0.1
Linoleic Acid (trans-9, trans-11), %	4.4	4.8	4.3	1.2	1.4	1.6	5.8	23.3	8.4	0.4
Eicosadienoic Acid, %	3.2	2.1	1.1	0.5	0.5	0.5	2.9	7.4	4.7	—
Eicosatrienoic Acid, %	2.6	2.9	1.9	0.4	0.6	0.5	6.4	23.6	9.8	—
Behenic Acid, %	0.4	0.9	0.8	0.7	—	0.7	2.6	5.2	0.5	—
Higher Boiling Acids, %	—	—	—	—	—	—	—	—	0.9	—

[a] Courtesy of Arizona Chemical Company, Wayne, New Jersey

Table 47. Typical Analysis of ACINTOL Tall Oil Fatty Acids and Selected ACINTOL Distillation Products[a]

Property	D6LR	FA-1	FA-1 Special	FA-2	FA-3	EPG	D29LR	DLR Special	Heads 2122	Tall Oil Pitch
							ACINTOL DISTILLATION PRODUCTS			
Color, Gardner	10	5	3	1–2	2	1–	5	5+	16	12–15
Acid Value	192	195	197	197	199	198	198	191	140–160	22–55
Saponification Value	196	197	198	199	200	200	193	196	—	100
Iodine Value (Wijs)	135	131	131	130	130	130	—	—	—	—
Composition										
Moisture, %	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	0.1	0.1
Ash, %	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	0.08
Rosin Acids, %	6.5	4.2	2.5	0.9	0.4	0.5	29	23	0.6	15–25
Unsaponifiables, %	2.7	1.6	1.6	0.9	0.5	0.5	2	1.4	25	30–36
Fatty Acids Total, %	90.8	94.2	95.9	98.2	99.1	99.0	69	75.6	74.4	34–50
Fatty Acid Composition: Polyunsaturated, Conjugated, as Linoleic, %	—	8	7	6	5	5	—	15	1	—
Polyunsaturated, Nonconjugated, as Linoleic, %	—	32	34	36	38	38	—	25	18	—
Oleic, %	—	44	46	47	49	49	—	35	20	—
Saturated, %	—	5	4	3	3	2	—	4	35	—
Others, By Difference, %	—	11	9	8	5	6	—	21	26	—
Softening Point	—	—	—	—	—	—	—	—	—	35
Specific Gravity, 25°/25°C	0.910	0.906	0.902	0.898	0.897	0.897	0.943	0.931	0.920	1.011
Weight Per Gallon, 25°C, lbs	7.57	7.53	7.50	7.47	7.45	7.45	7.85	7.74	7.60	8.42
Viscosity, Gardner-Holdt, 25°C	A	A	A	A	A	A	D	B	—	—
Viscosity, SUS, 100°F	120	100	97	94	95	93	—	—	—	—
Viscosity, SUS, 210°F	—	—	—	—	—	—	51	47	40	52
Flash Point, Open Cup, °F	410	380	390	400	420	400	400	400	370	520
Fire Point, Open Cup, °F	445	415	415	435	450	435	440	437	400	545

[a] Courtesy if Arizona Chemical Company, Wayne, New Jersey

Table 48. Tall Oil Esters (161)

Commercial Name of Product	Produced by	Acid Number Min.	Acid Number Max.	Iodine Number Min.	Iodine Number Max.	Saponification Number Min.	Saponification Number Max.	Specific Gravity 15.5° C./15.5° C. Min.	Specific Gravity 15.5° C./15.5° C. Max.	Refractive Index at 25.0° C.	Color Gardner 20°-30° C.	Absolute Viscosity at 25° C. Gardner-Holdt	Absolute Viscosity at 25° C. Poises	Notes
★ Albatol 300	Albemarle		13						1.00		11–12	Z3		Penta ester. 59% oil.
★ Albatol 600	Albemarle		13						1.00		11–12	Z6		Penta ester. 59% oil.
★ Albatol 3480	Albemarle		13						0.955		11–13	Y-Z		Penta-maleic ester. 80% nonvolatile.
☆ Flex-Synol 105 Z-6	Am. Alkyd	6	12					0.98	0.99		9 –12	Z6		Oil extended.
Esterified Tall Oil	Brown	Processed to desired specifications.												
#156, Esterified Tall Oil	Calif. Flax	8.0	14.0								12–14			Appearance hazy.
#766, Esterified, Extra Pale	Calif. Flax	8.0	12.0								10–12			Appearance brilliant.
Modiphat 145	General	90	100			162	170				12–13			Semi-solid. Partially esterified tall oil.
Modiphat 245	General		9.0			155	165				Amber			Semi-solid. Pentaerythritol ester, of tall oil.
Modiphat 245-CWO	General		7.0								9 max	Z2-Z4		Blend of equal parts tung oil and tall oil ester.
Dryfol MP	Smith		5.0					0.918			12	W-X		Nonvolatile, 60%. Penta maleic ester of refined tall oil.
Smithco "PE"	Smith		15.0	170	175	130	136	1.000	1.010		10–12	Z5-Z6		Pentaerythritol ester of tall oil.
Smithco "RT"	Smith	6.0	10.0	170	176	130	136	1.000	1.010		11–12	Z5		Glycerine ester of refined tall oil.
★ Petal Ester	Synthetic	10	15								9 –11	Z6+		Penta ester
☆ Glycertal Ester	Synthetic	5	10								9 –11	Z5-Z6		Glyceryl ester
★ Aroplaz 1119	U.S.I.	6	11					0.980	0.990		12–14	Y-Z1		100% solids
★ Aroplaz 1129-M	U.S.I.	12	18					0.956	0.967		12–14	O-W		80% solids in mineral spirits

Table 49. Tall Oil Fatty Acid Esters[a]

	TYPICAL ANALYSIS				
	CROPLAS MX	CROPLAS BX	CROPLAS TH	CROPLAS EH	CROPLAS IO
Specific Gravity @ 25 C	0.8825	0.9145	0.9303	0.8708	0.8703
Refractive Index @ 20 C	1.4585	1.4593	1.4705	1.4598	1.4600
Color, Gardner	4	4	4	2	3
Saponification Value	189	165	153	141	140
Iodine Value	117	105	95	90	89
Unsaponifiables %	2.0	2.0	2.0	2.0	2.0
Acid Value	0.2	0.5	0.7	0.2	0.3
Rosin Acids %	0.1	0.3	0	0	0.1
Moisture %	Nil	Nil	Nil	Nil	Nil
Weight/gallon, pound	7.36	7.6	7.76	7.27	7.25

[a] Courtesy of Crosby Chemicals, Inc., Picayune, Mississippi

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